Emission channeling from short-lived isotopes

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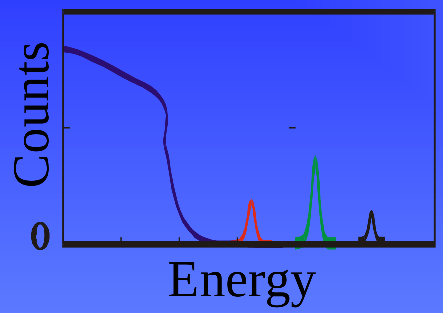
- ❑ **The emission channeling technique**
  - **Basic principles**
  - **History**
  - **Experimental requirements + setups**
- ❑ **Examples of interest to a MYRRHA based ISOL facility:**
  - **Lattice site changes and diffusion of Li in compound semiconductors**
  - **Lattice site changes of Na in ZnO**
  - **Lattice sites of rare earths in GaN**
- ❑ **Conclusions**

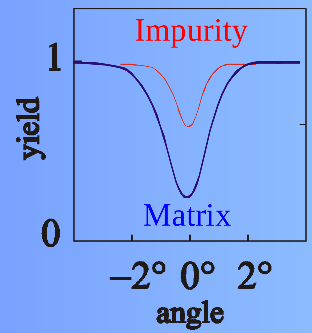
Emission channeling *vs* ion beam channeling



If you want to know...

- sample stoichiometry
- multilayer thicknesses
- impurity depth profiles

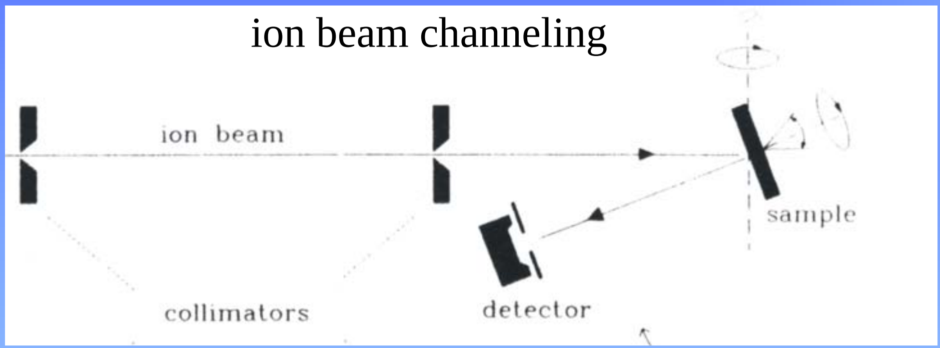
⇒ use of Rutherford Backscattering (RBS)



If you need...

- profile of lattice defects
- impurity lattice site
- impurity rms displacements

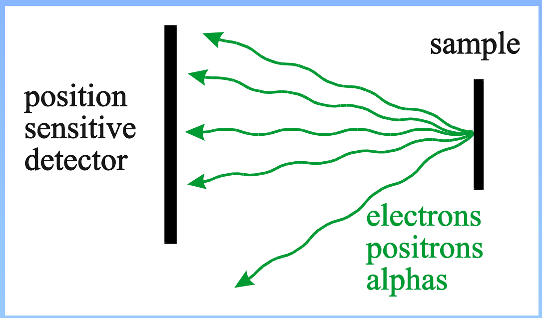
in **single crystals**



...then you can use ion beam channeling with RBS, PIXE or NRA

<u>but</u>:

requires impurity doses above ~10^{14} cm^{-2} .



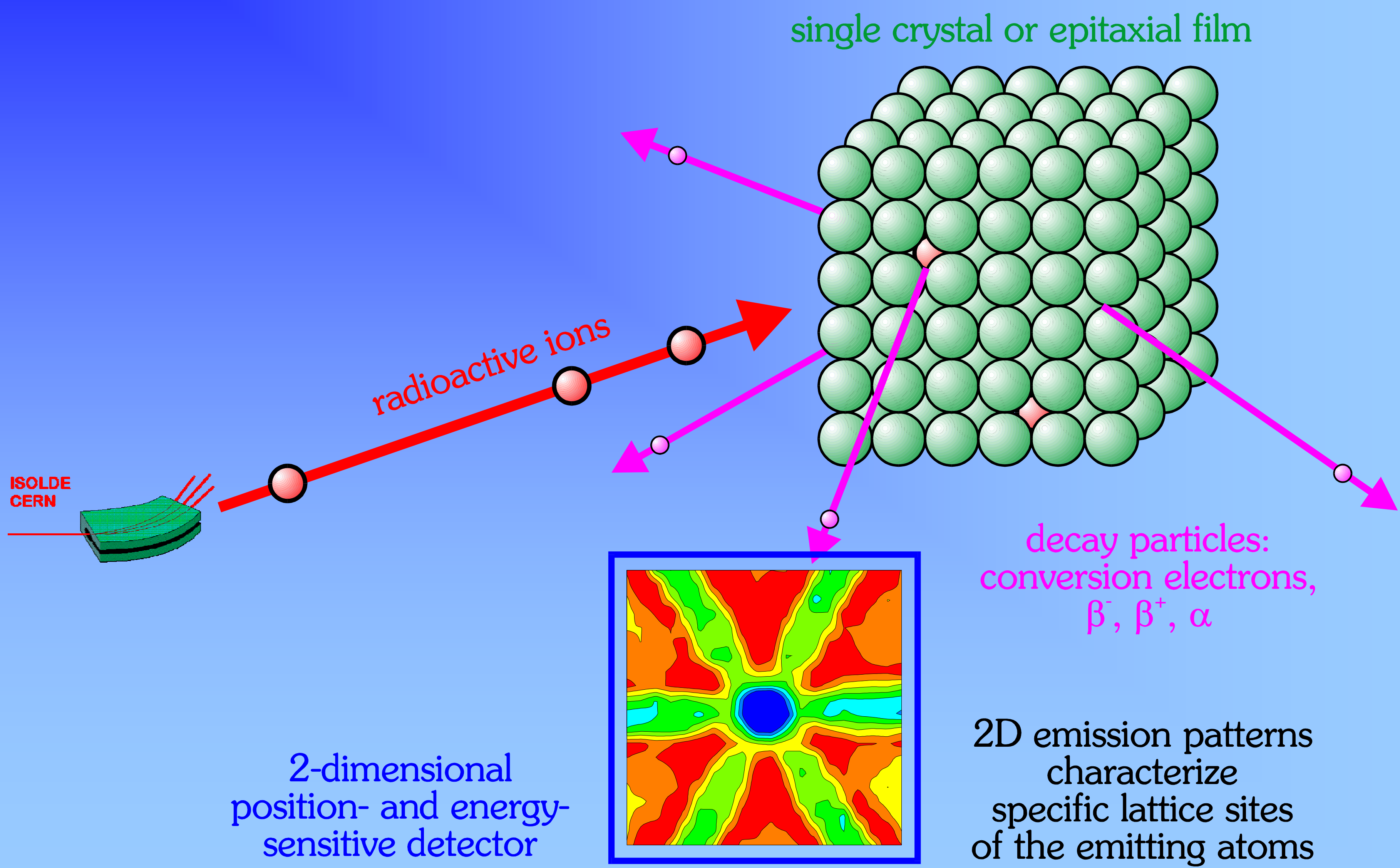
If you ask for...

lattice location at low fluences (10^{12}-10^{14} cm^{-2})

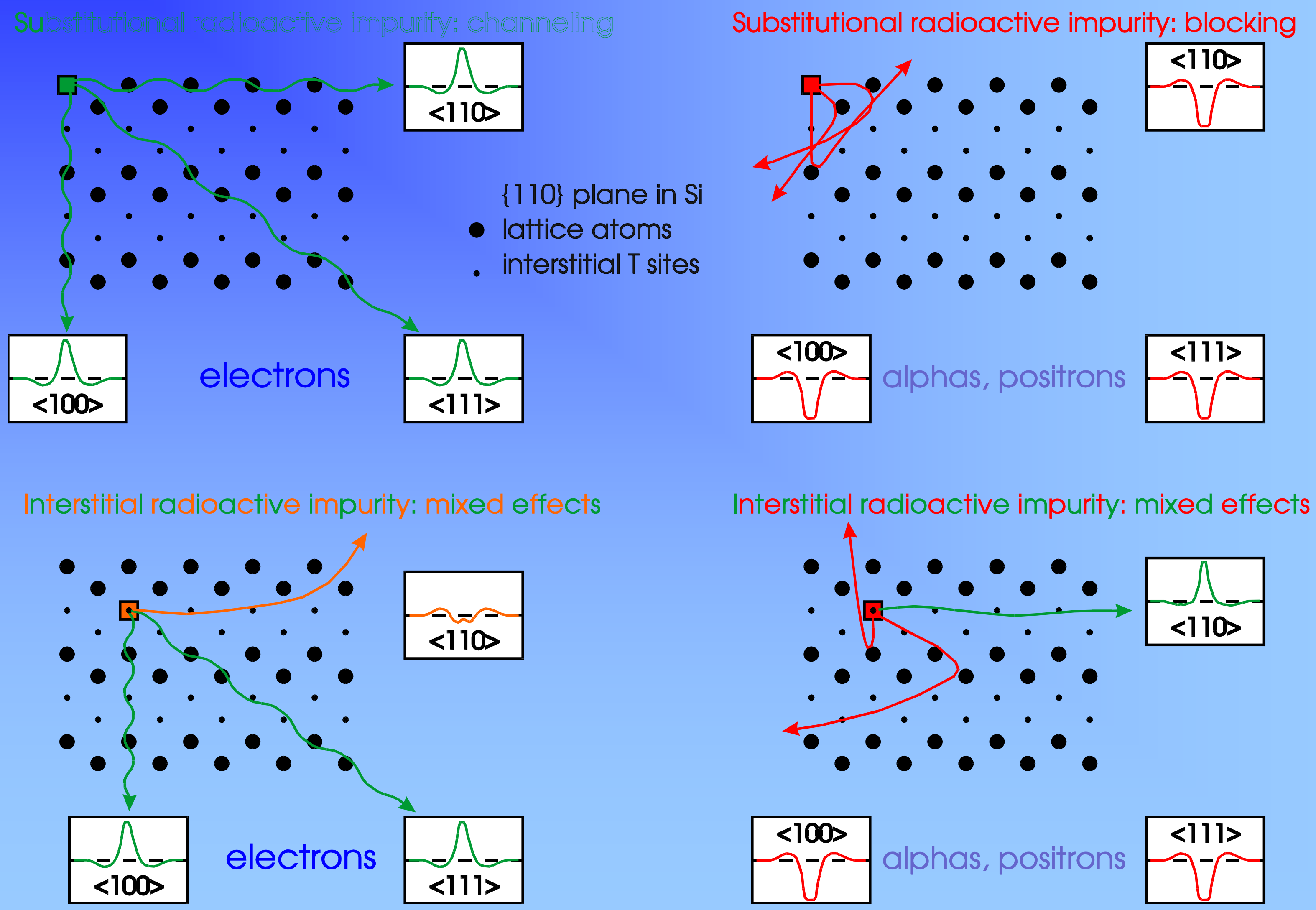
⇒ use **radioactive impurities: emission channeling**

- ~ 4 orders of magnitude higher efficiency than RBS
- independent of mass of impurity
- no damage from probing beam

Emission channeling lattice location of radioisotopes



Characteristic differences between electron and alpha (or positron) emission channeling



Computational approaches to simulate emission channeling

- Alpha emission channeling:
  theoretical description identical to ion beam channeling i.e. is best done by Monte Carlo methods (calculating many ion trajectories). The "method of choice" but unfortunately not many α emitting isotopes exist.

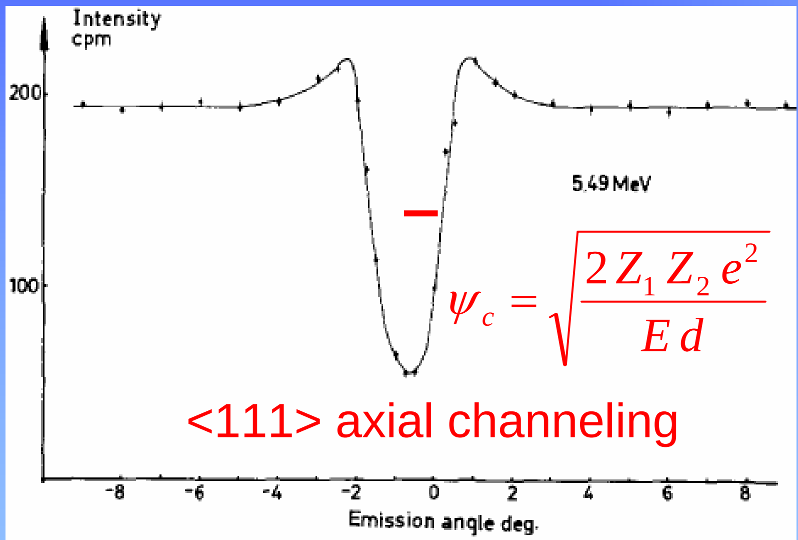
- Positron emission channeling:
  theoretical description exists (either Monte Carlo or quantum-mechanical) but not well-adapted to experiments. Number of positron emitters much smaller than electron emitters.

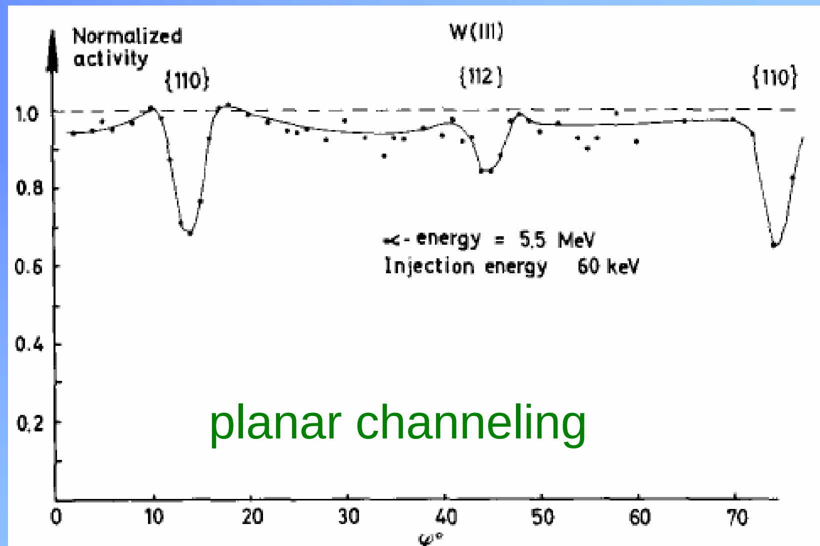
- Electron (β^- and CE) emission channeling:
  well-developed method, accounts for the majority of experiments. Theoretical description by means of "manybeam" formalism = quantum-mechanical approach, solution of Schrödinger equation in a periodic potential.

Emission channeling history

The first lattice location experiment by means of charged particles, done at Royal Institute of Technology, Stockholm,
B. Domeij and K. Bjørkvist: "Anisotropic emission of α-particles from a monocrystalline source", Physics Letters 14 (1965) 127-128.

Angle-dependent α emission rate from ^{222}Rn implanted into W

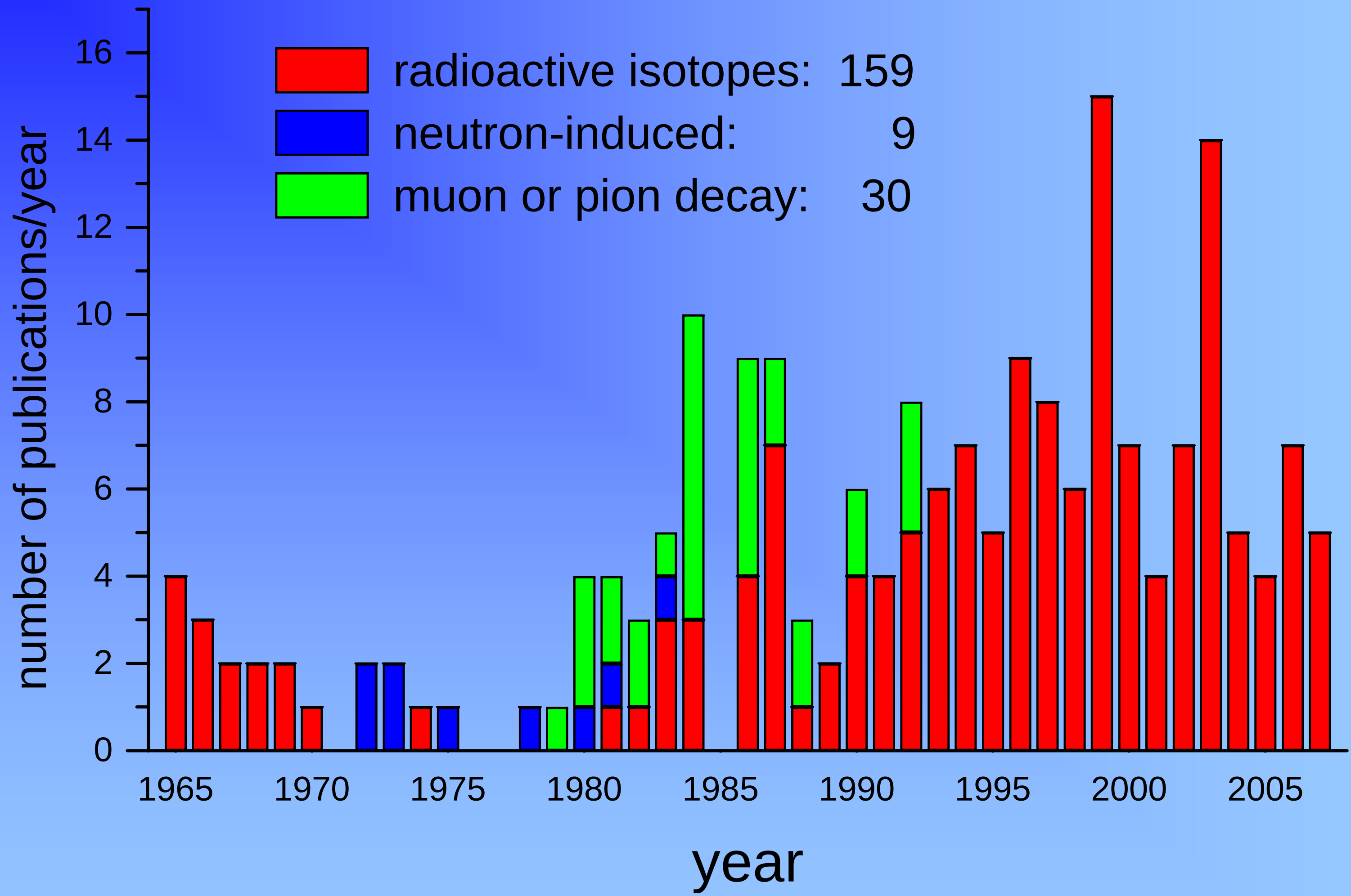




⇒ implanted Rn occupies substitutional W sites

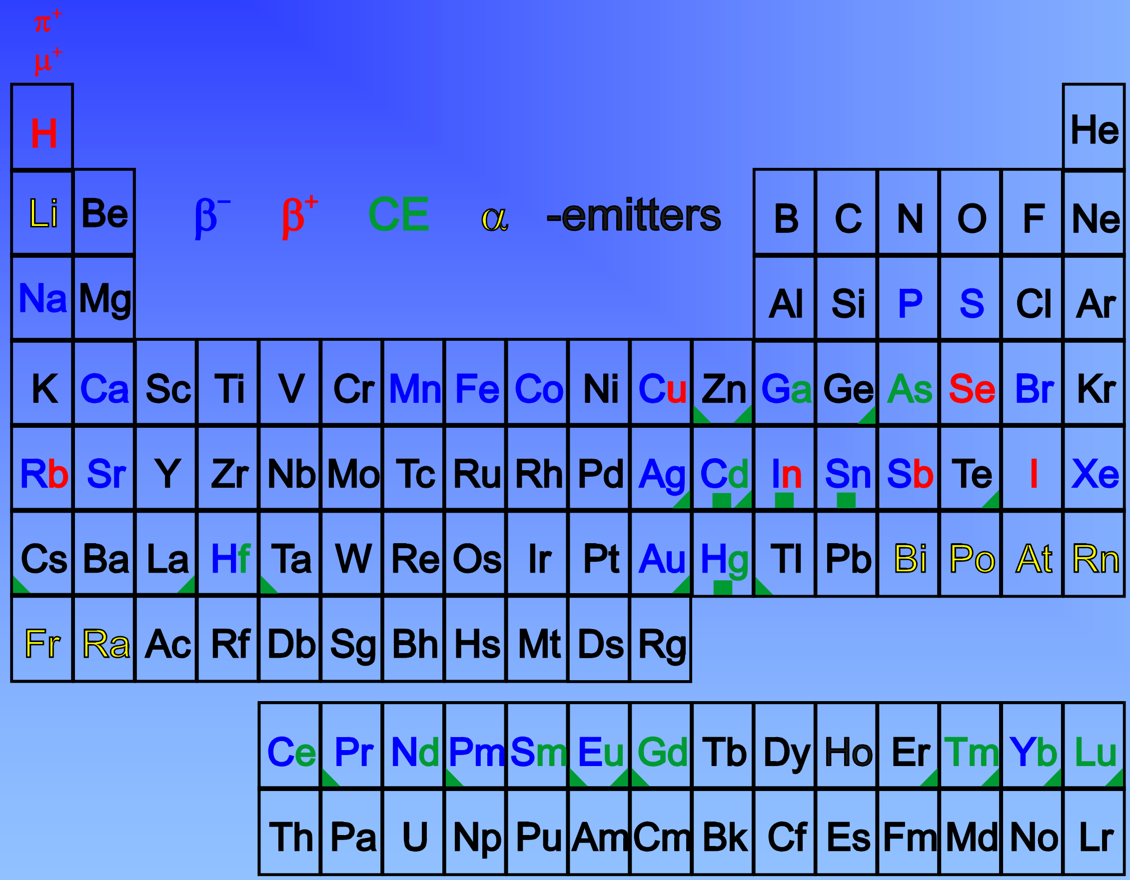
Emission channeling lattice location is in fact older than RBS channeling!

Emission channeling publication history



Experiments with radioactive isotopes (produced and implanted mostly at ISOLDE/CERN) account for the majority of emission channeling experiments

Elements for which emission channeling experiments have been reported



β^-, β^+, **CE** or α emitting isotopes exist for most elements of the periodic system

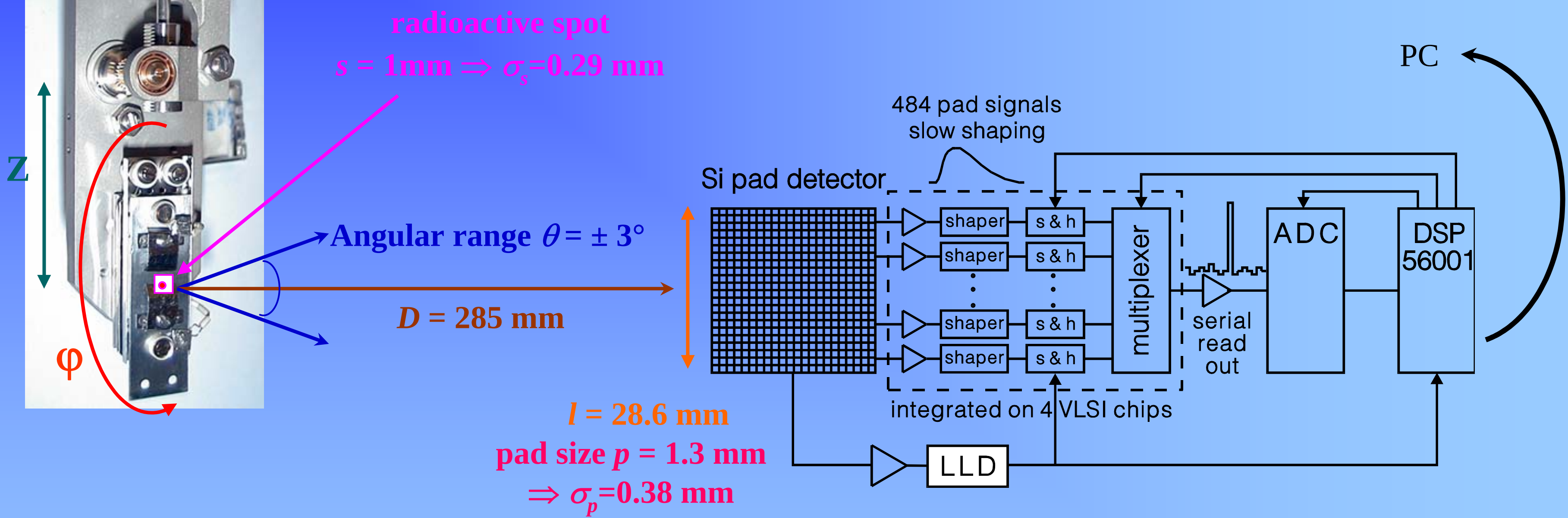
⇒ many experimental possibilities

<u>Important criteria for suitable isotopes:</u>

• half life of decay • energy of emitted particles • availability as ion beam
• possible superposition from decay chains • radiation protection issues

Experimental details - geometry

Angular width $\approx \psi_c = \sqrt{\dfrac{2 Z_1 Z_2 e^2}{E\, d}}$ = 0.9° for 150 keV e^- in Si <100>



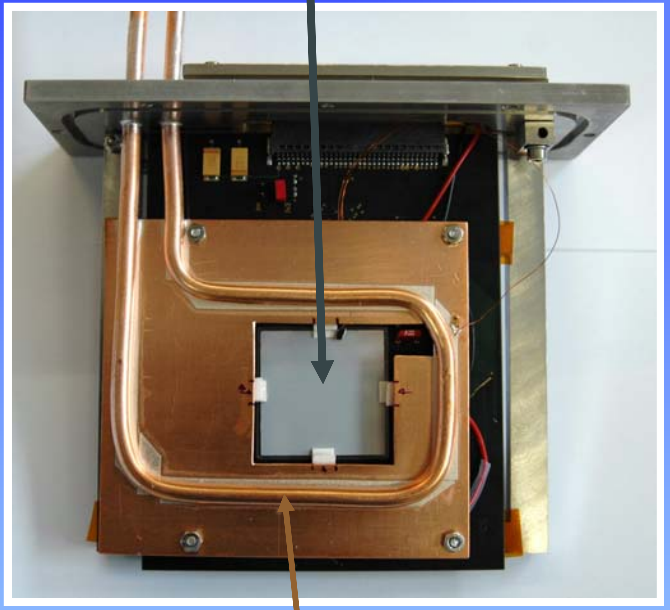
Angular resolution $\Delta\theta$ depends on both size of radioactive spot σ_s and detector resolution σ_p :

$$\Delta\theta \approx \arctan \frac{\sqrt{\sigma_s^2 + \sigma_p^2}}{D} = 0.1°$$

For a detector of size l the distance sample-detector D regulates both the angular range θ and the angular resolution $\Delta\theta$

Examples for position sensitive Si detectors

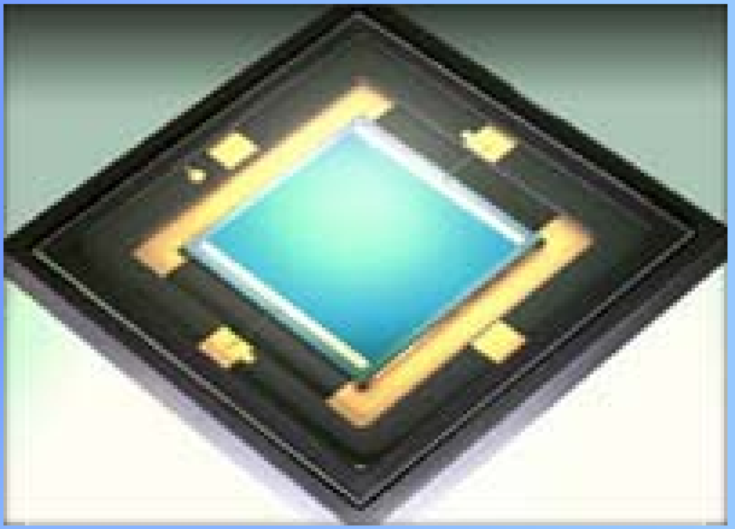
28×28 mm^2 22×22 pixel
Si pad detector for electrons



water & Peltier cooling (–30°C)

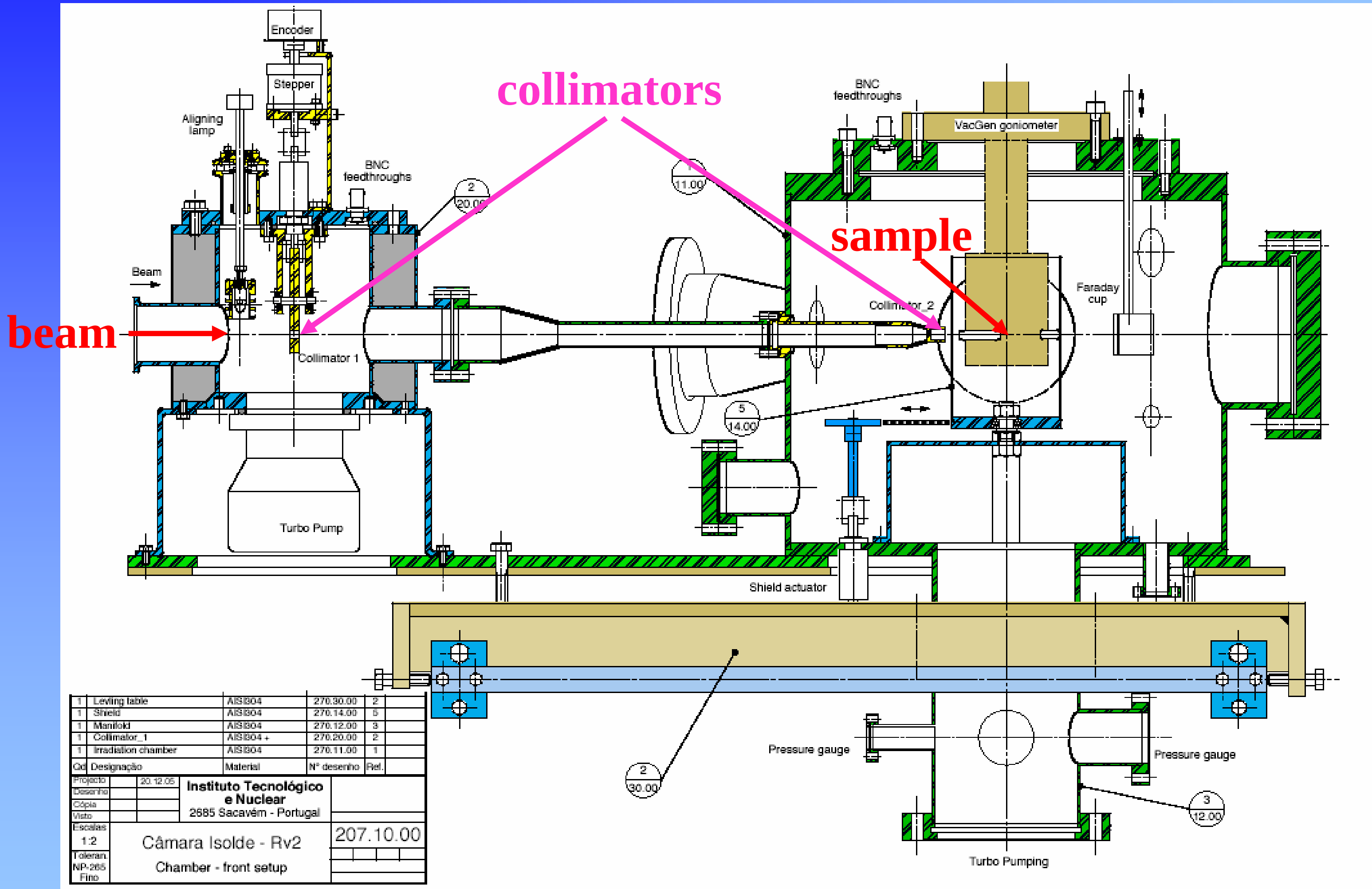
- prototype detectors (CERN)
- selftriggering pre-amplifier chips
- maximum count rate: ~3 kHz range
- energy resolution: photons ~1.2 keV
  electrons ~3 keV

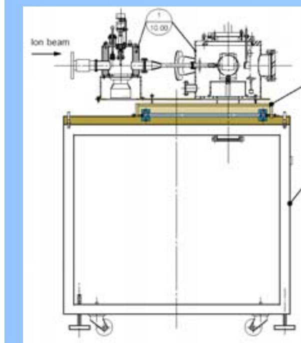
25×25 mm^2 resistive
charge division SiTeK detector
for α particles



- commercially available
- readout via conventional NIM modules + multiparameter ADCs
- maximum count rate: ~5-10 kHz range
- energy resolution: ~ 20-50 keV

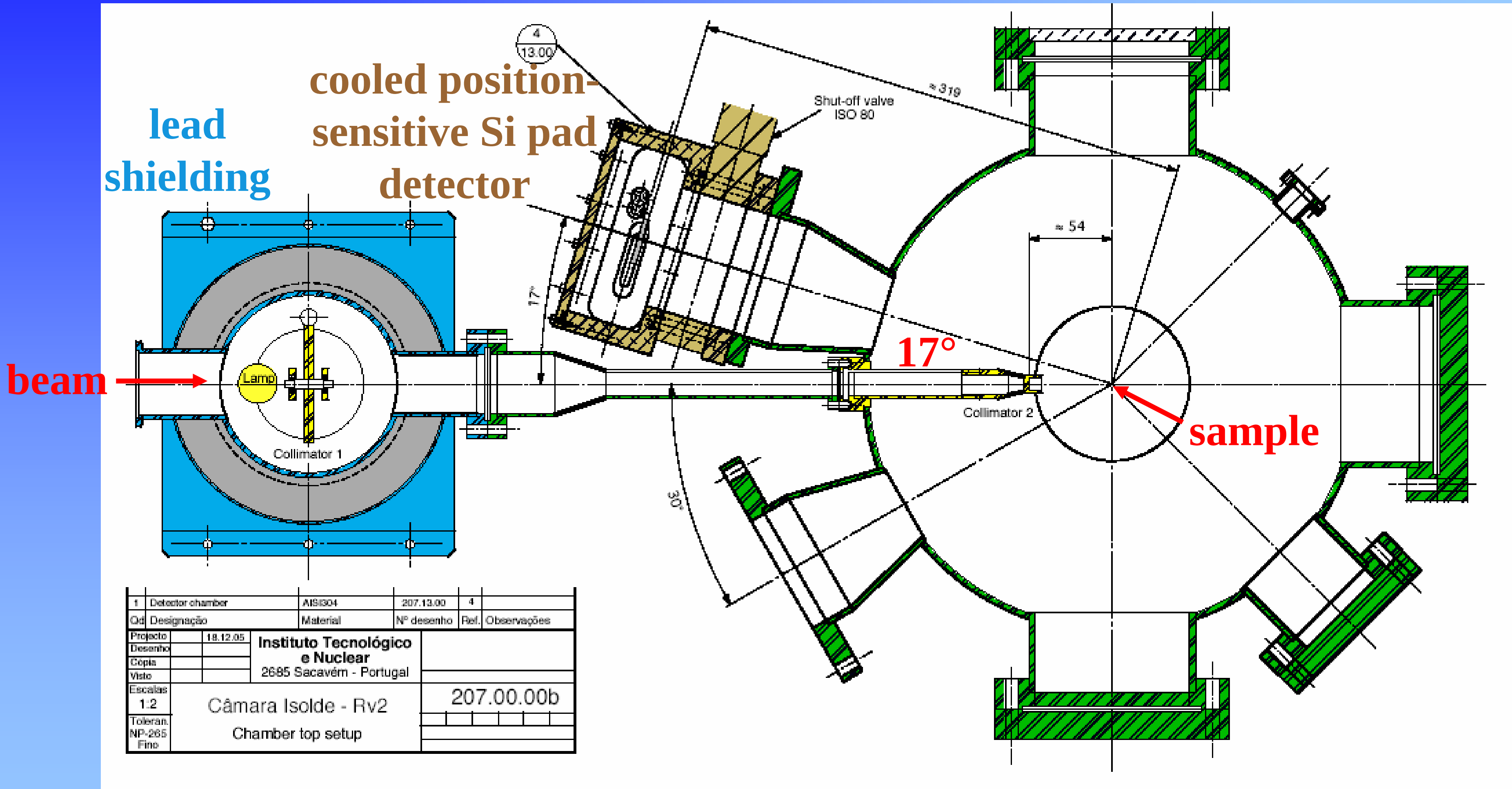
ITN on-line emission channeling setup: side view





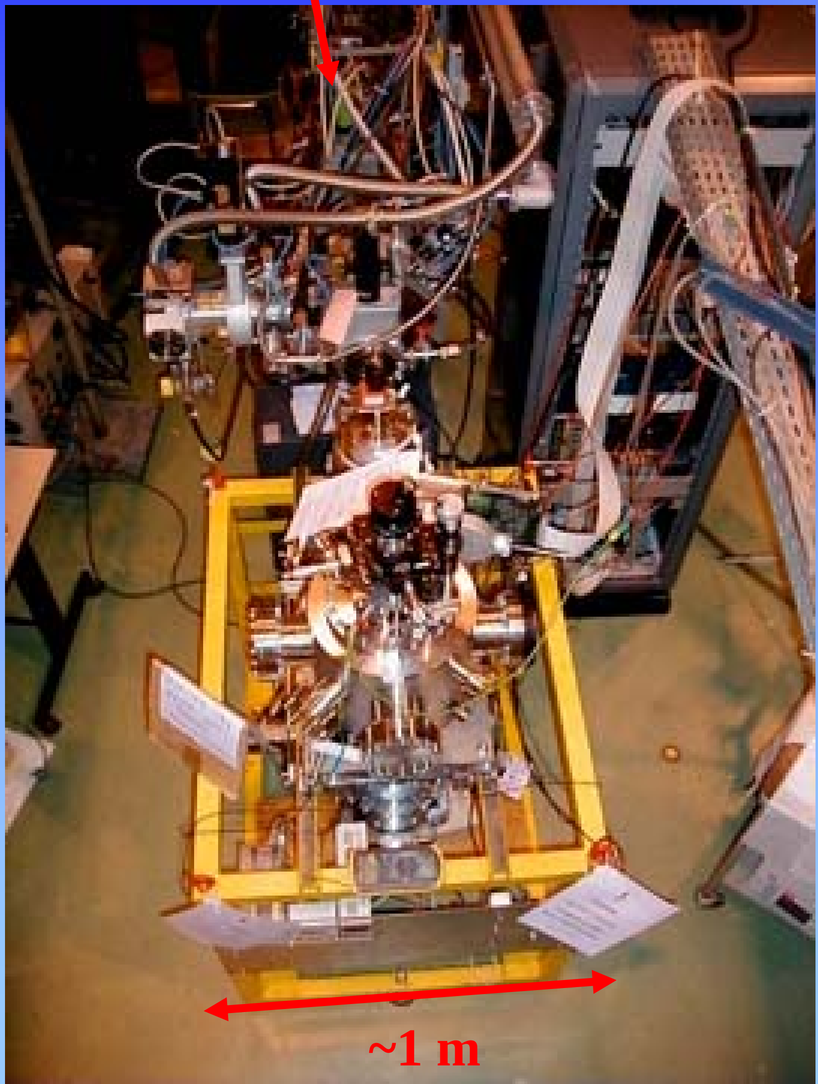
- ISOLDE beam is collimated by 2 apertures (1st variable size, 2nd ∅ 1 mm) on the sample
- sample mounted in 2- or 3-axis goniometer

ITN on-line emission channeling setup: top view

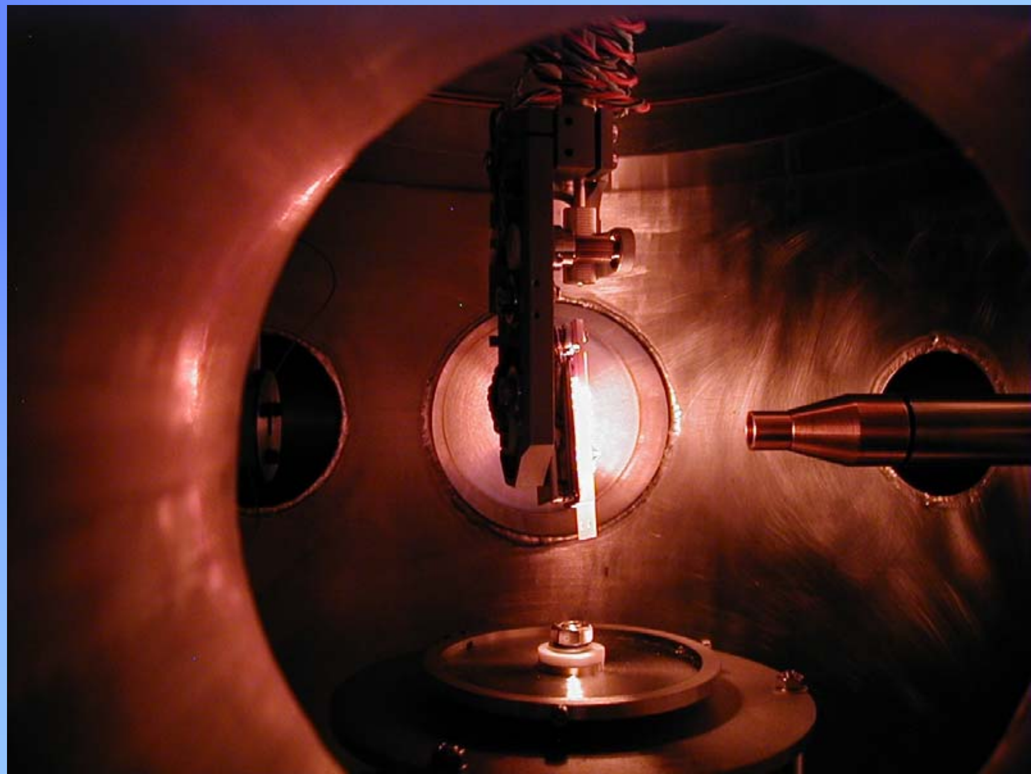


- detector at 17° backward geometry for simultaneous implantation and measurement
- valve in front of detector allows to maintain detector vacuum during sample exchange
- lead shielding around 1st collimator lowers background

New ITN on-line emission channeling setup



Inside view during 900°C sample annealing



Examples for experimental results

- α emission channeling:
lattice location of ^{8}Li in semiconductors

(diffusion processes of Li in semiconductors, Li-defect interactions)

- β^{-} emission channeling:
lattice location of ^{24}Na in ZnO

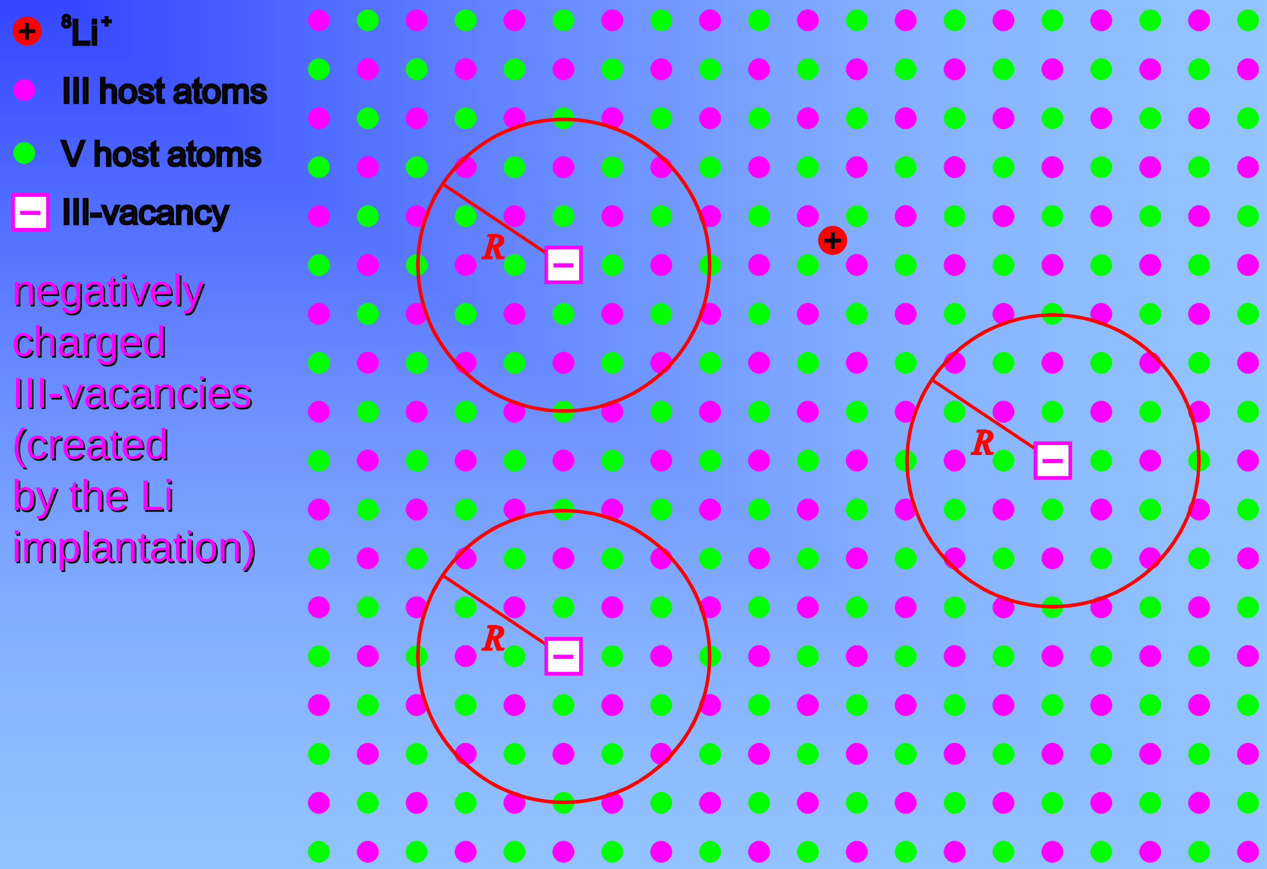
(interstitial Na_i donors *vs* substitutional Na_{Zn} acceptors)

- β^{-} and CE emission channeling:
lattice location of rare earths (REs) in GaN

(REs as optical or magnetic dopants)

Lattice site changes of implanted Li in III-V semiconductors

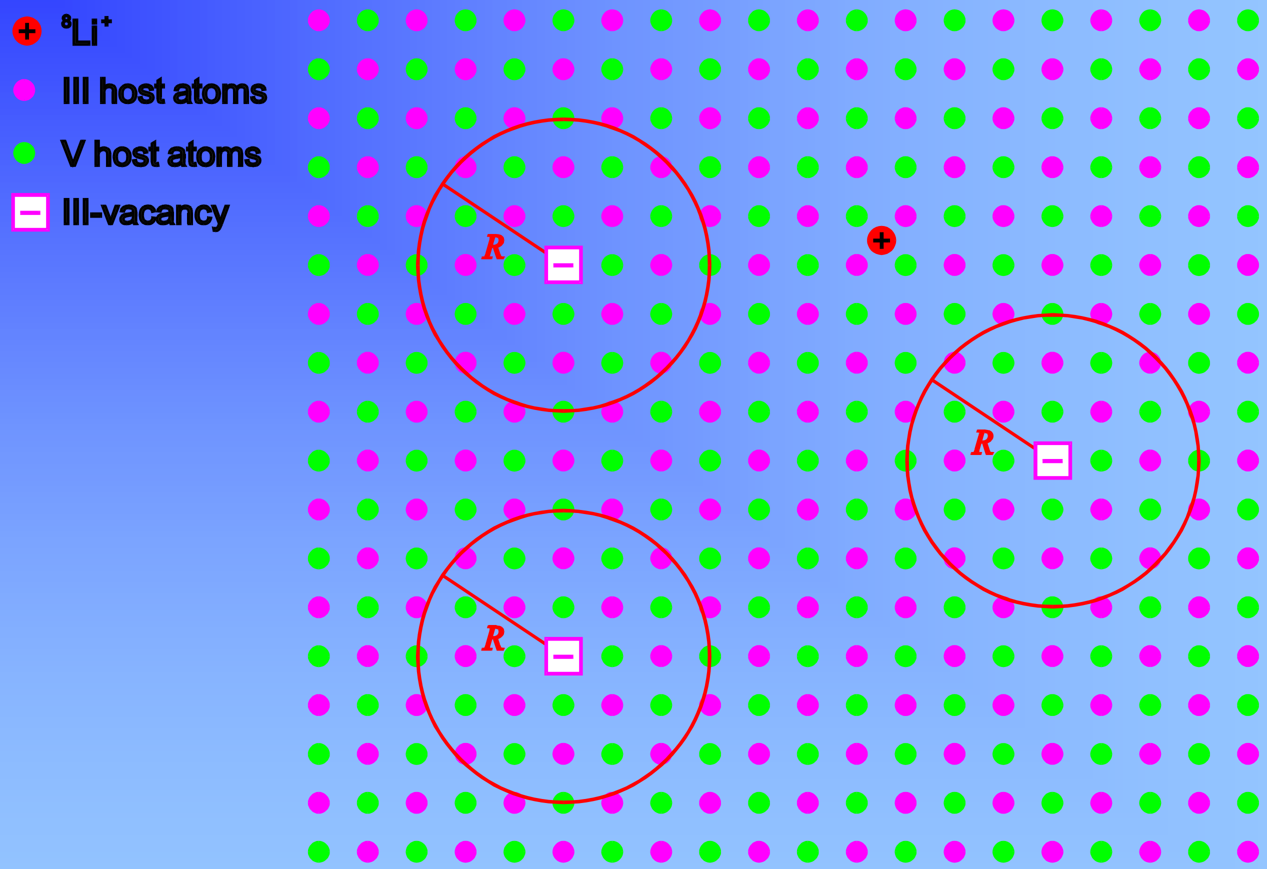
What is the fate of implanted Li in III-V semiconductors?



negatively charged III-vacancies (created by the Li implantation)

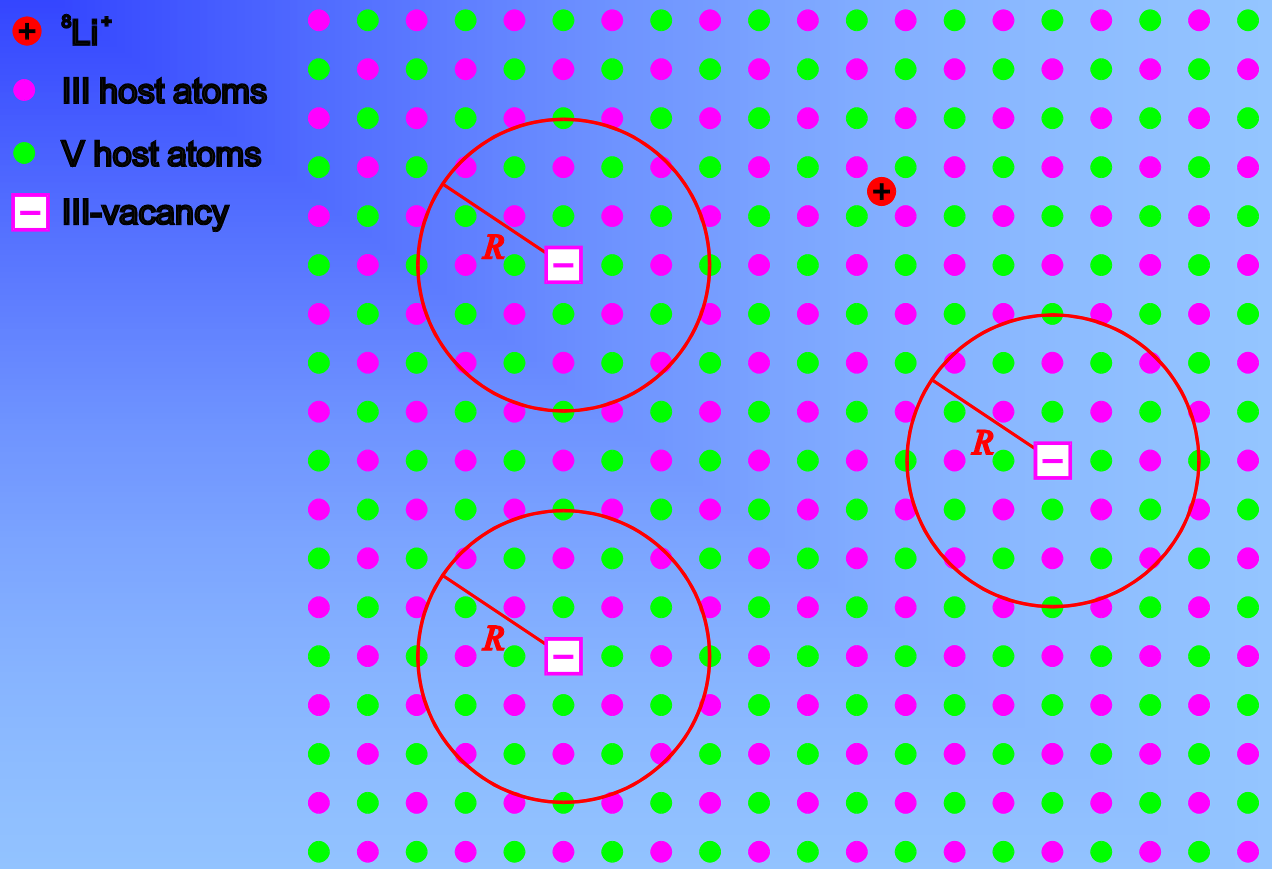
Low implantation temperature: Li is interstitial and immobile

Lattice site changes of implanted Li in III-V semiconductors



Increasing implantation temperature: interstitial Li starts to diffuse

Lattice site changes of implanted Li in III-V semiconductors



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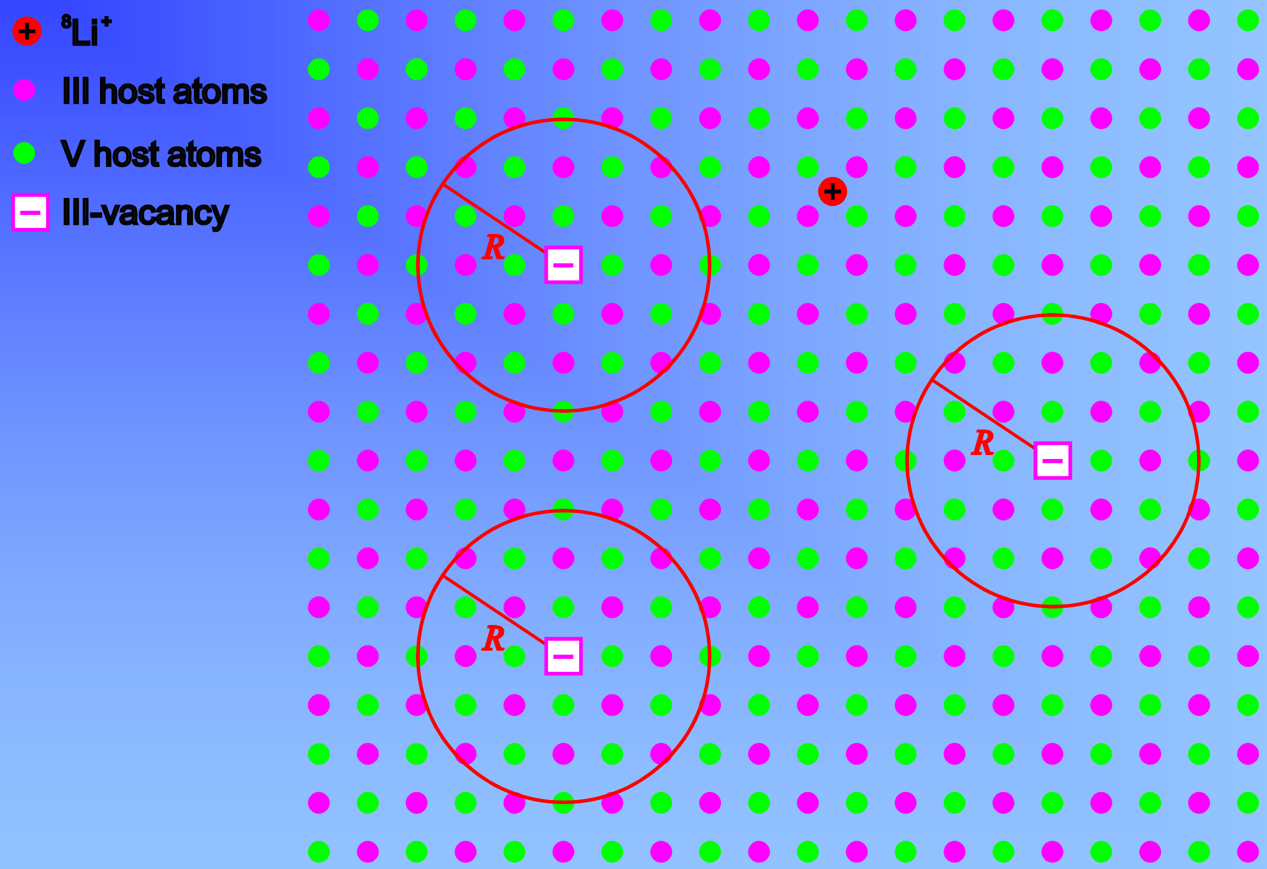

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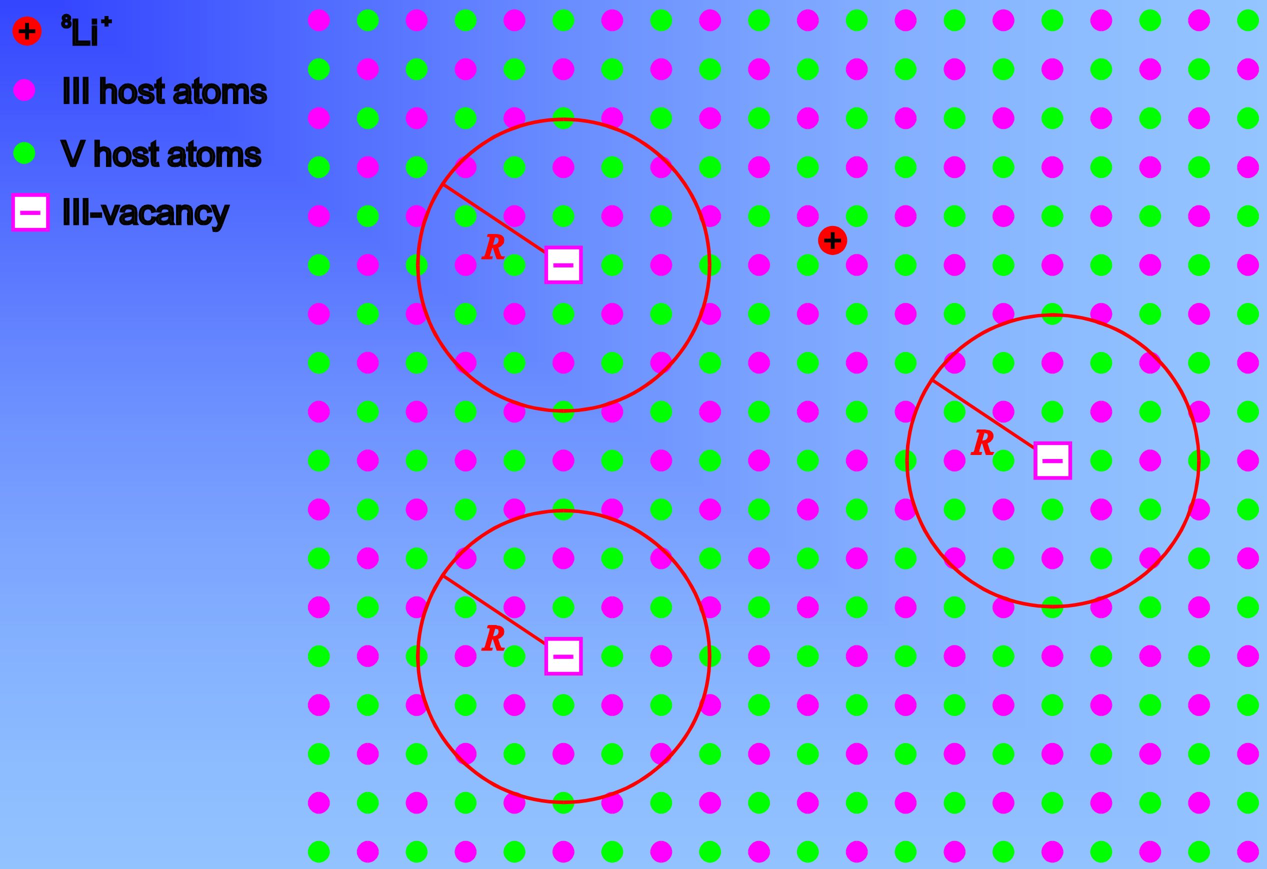

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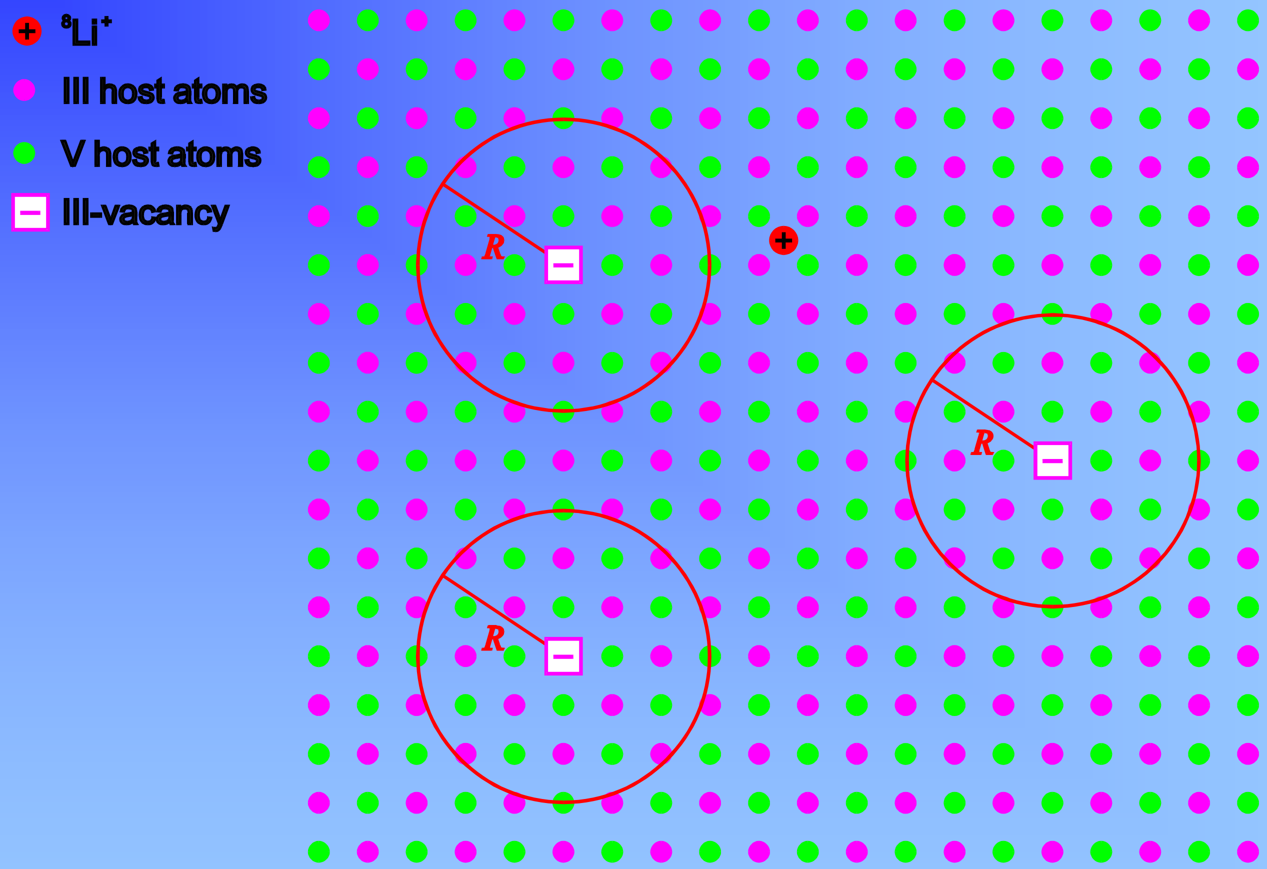

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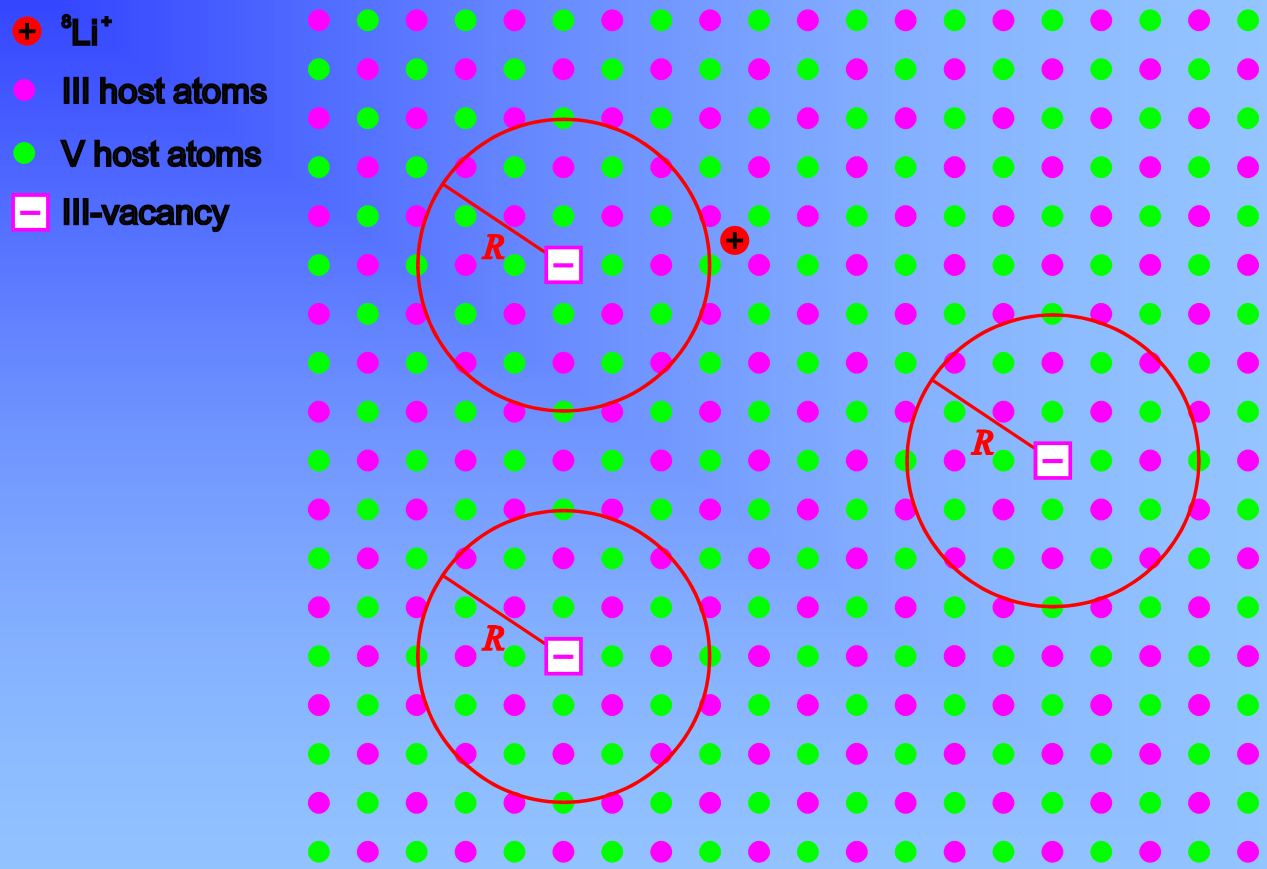

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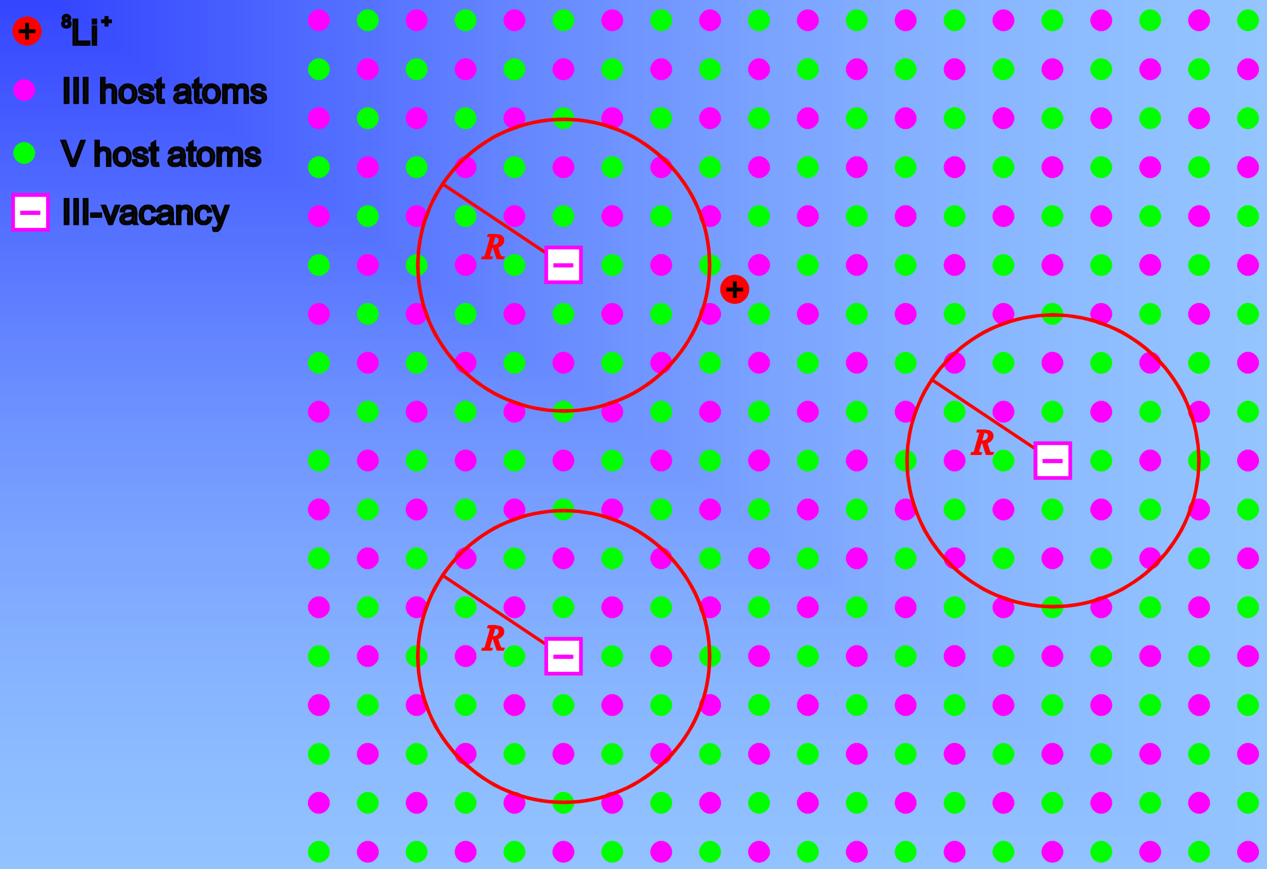

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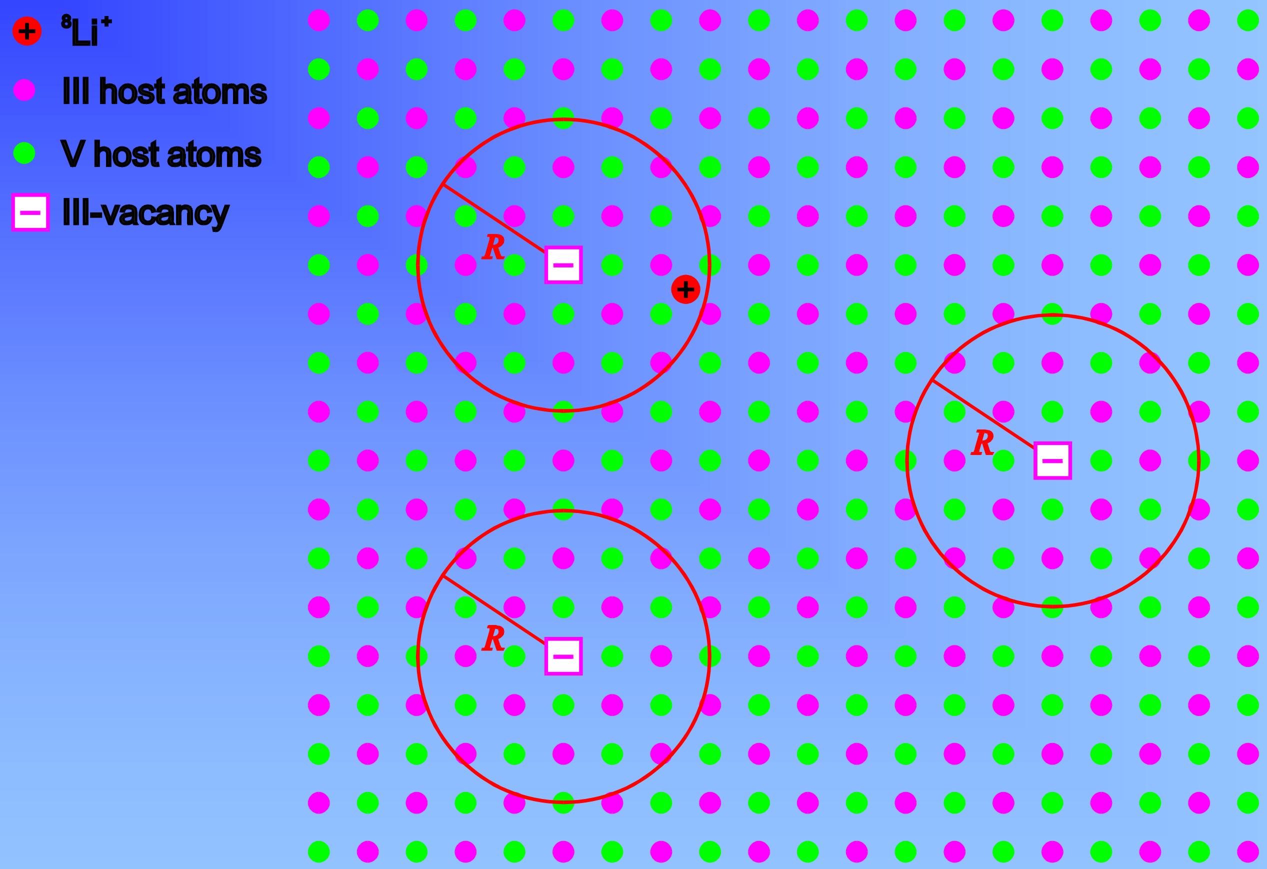
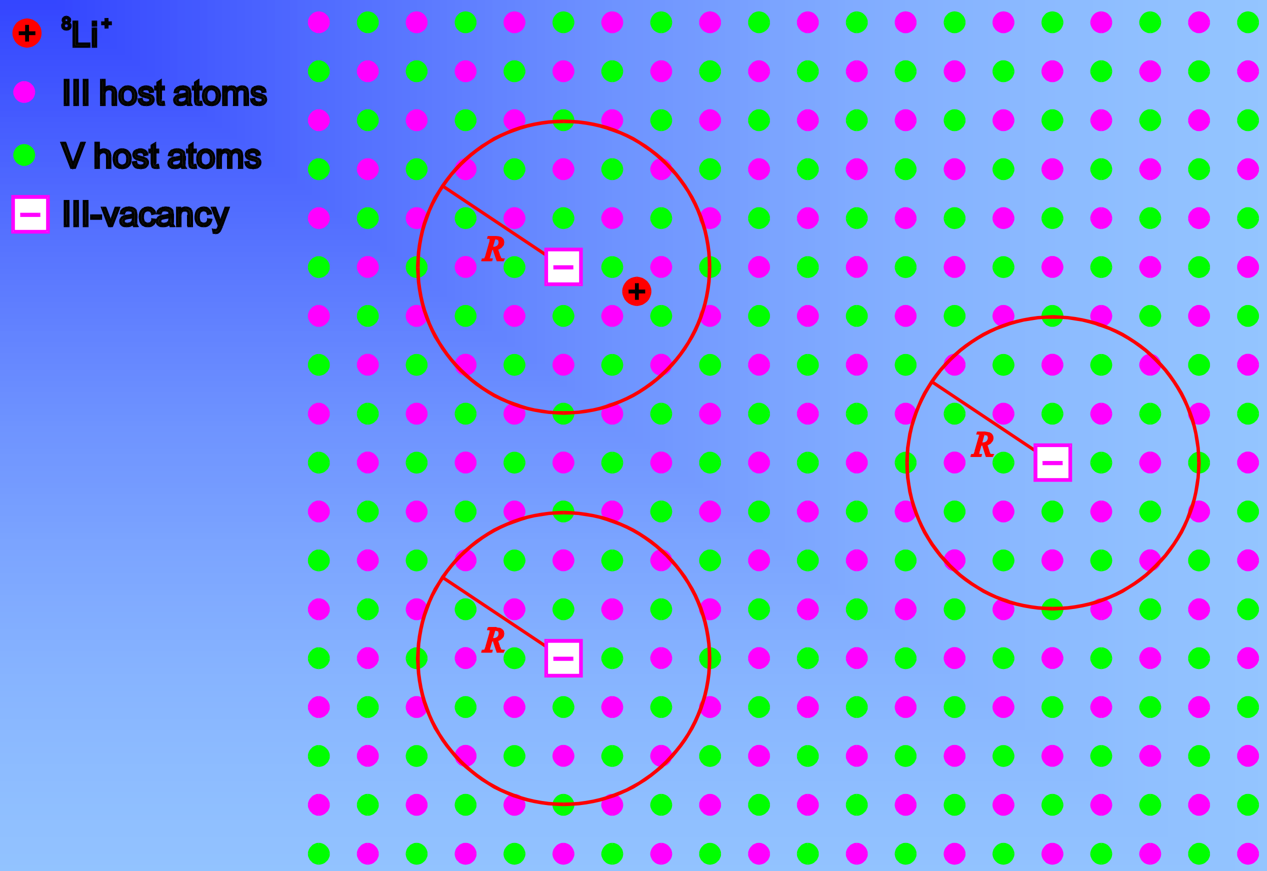


Having passed the Coulomb capture radius ***R***, Li^+ is attracted towards the negatively charged III-vacancy

Lattice site changes of implanted Li in III-V semiconductors



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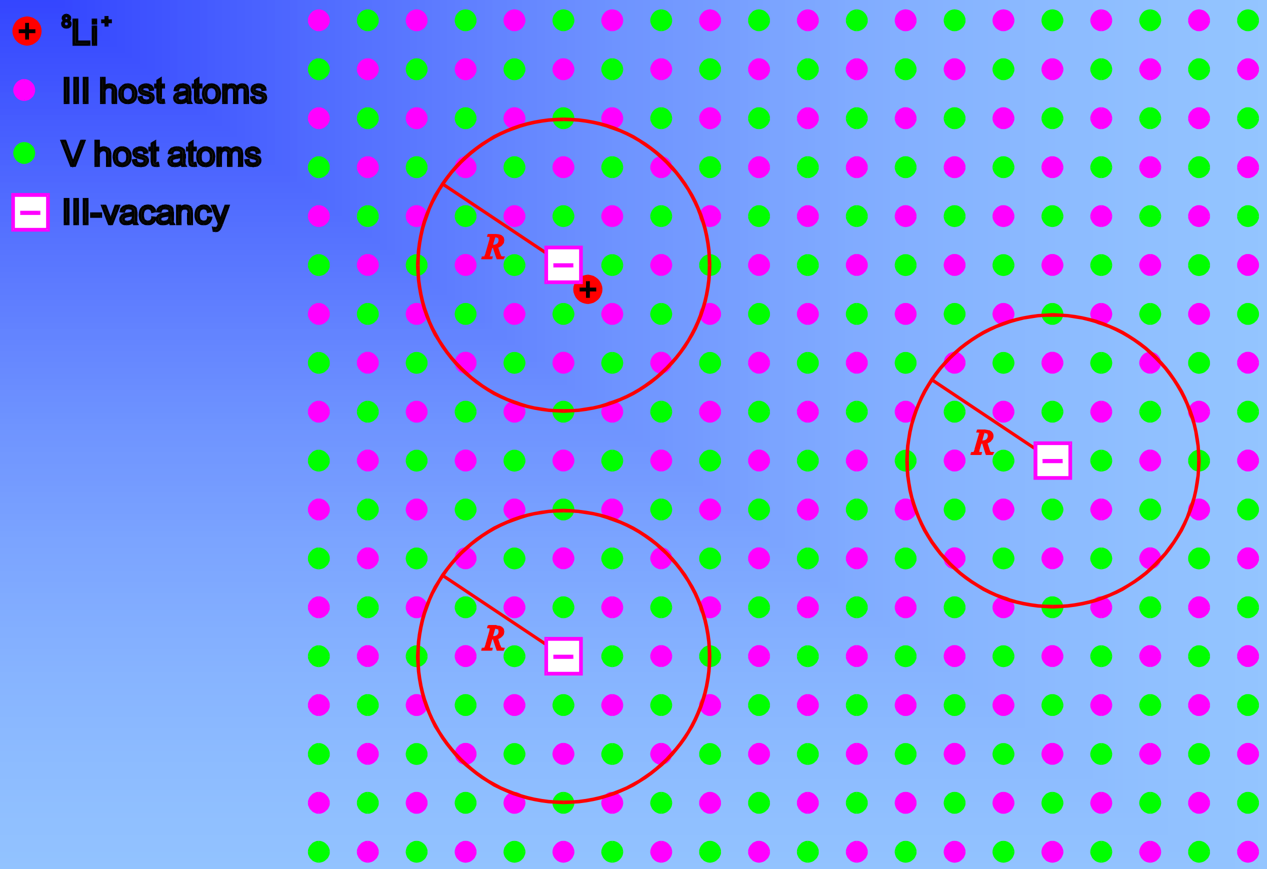
Lattice site changes of implanted Li in III-V semiconductors



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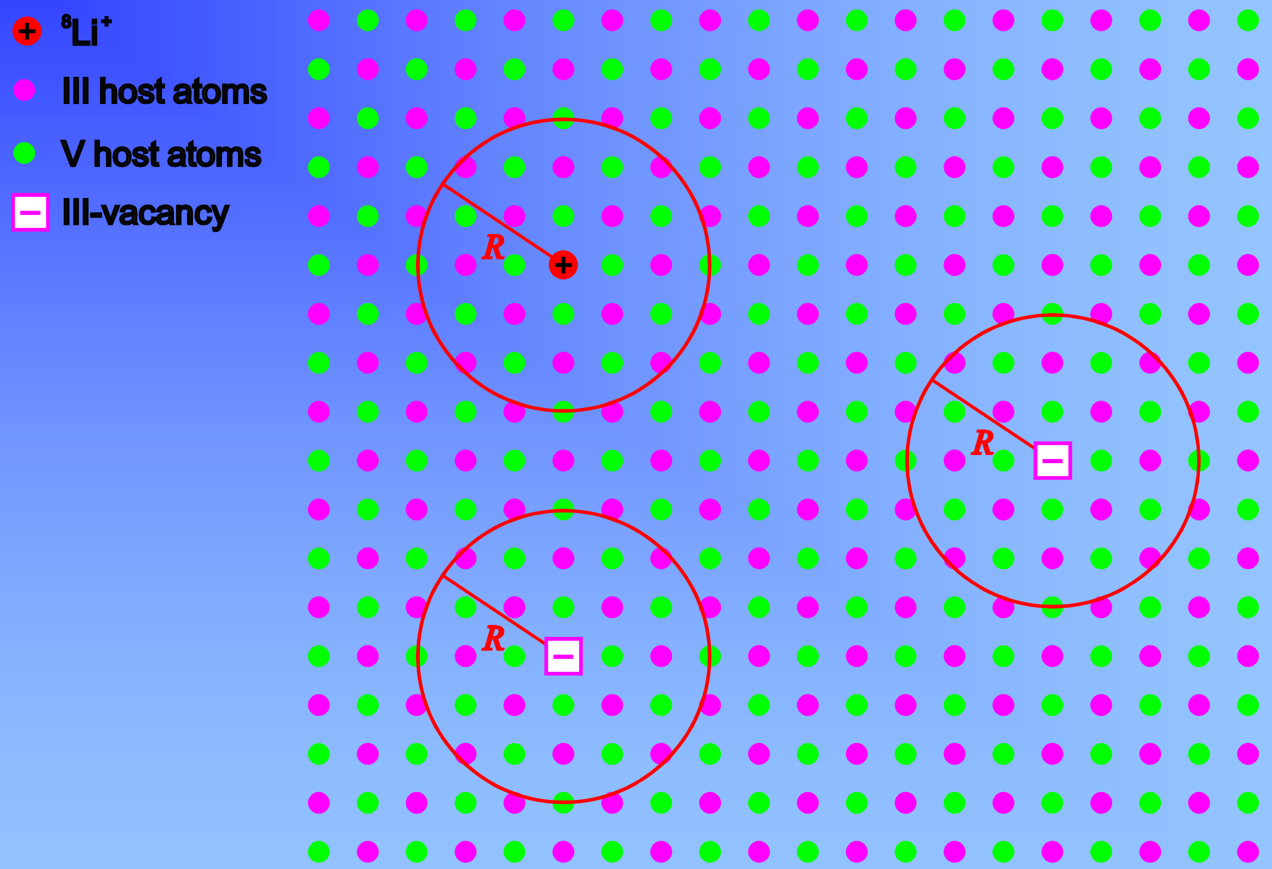
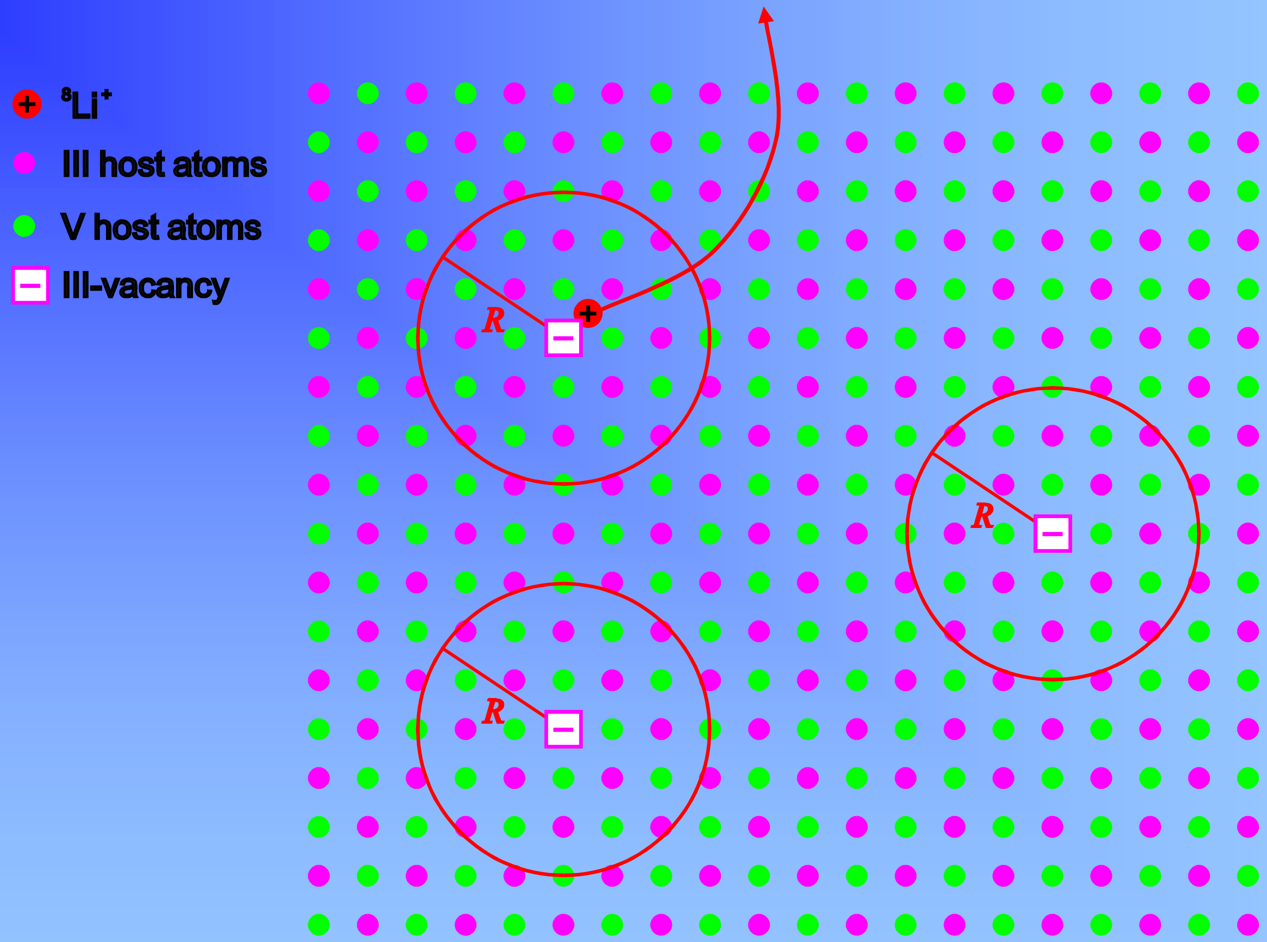


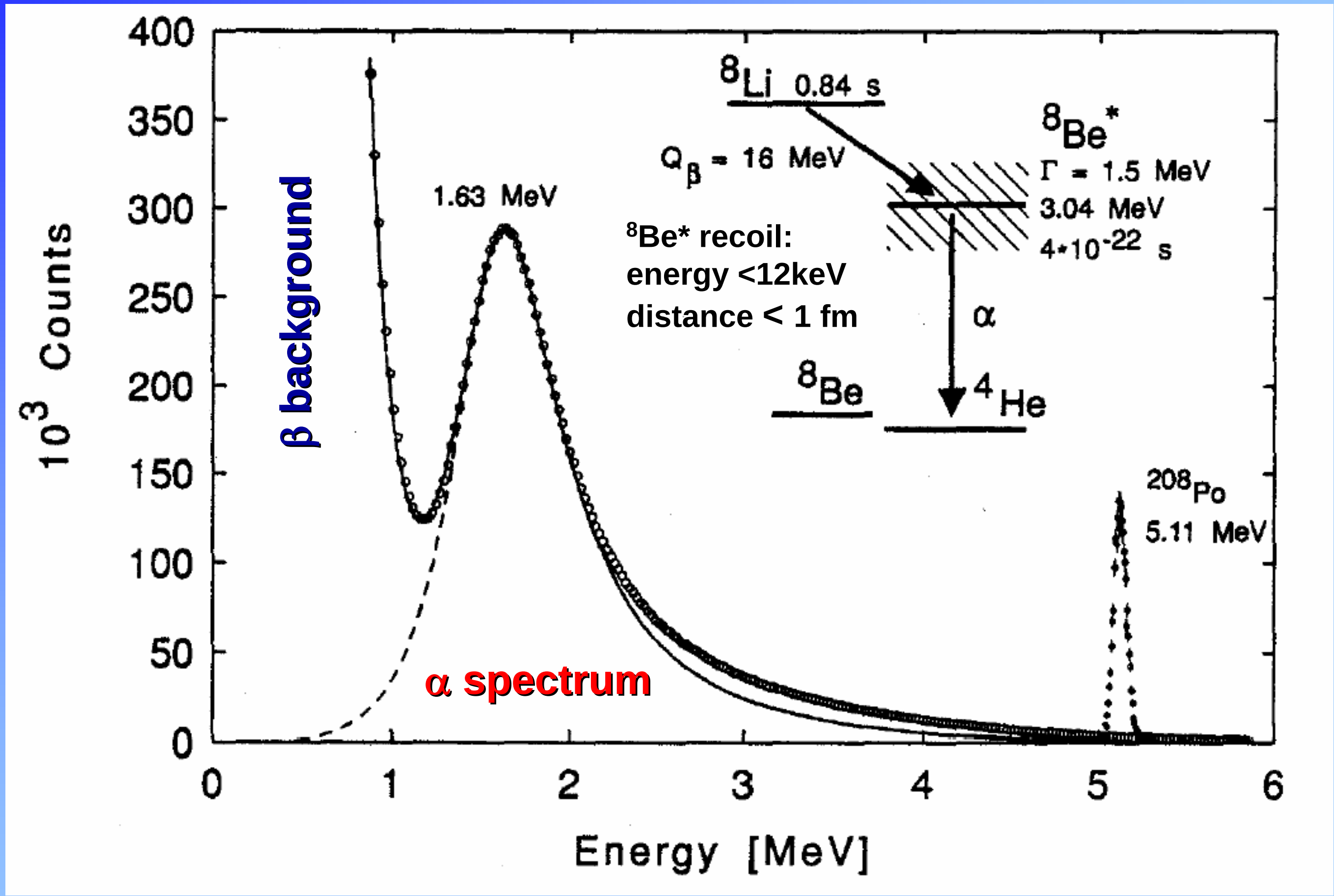
Li^+ finally combines with the III-vacancy, thus becoming substitutional

Lattice site changes of implanted Li in III-V semiconductors



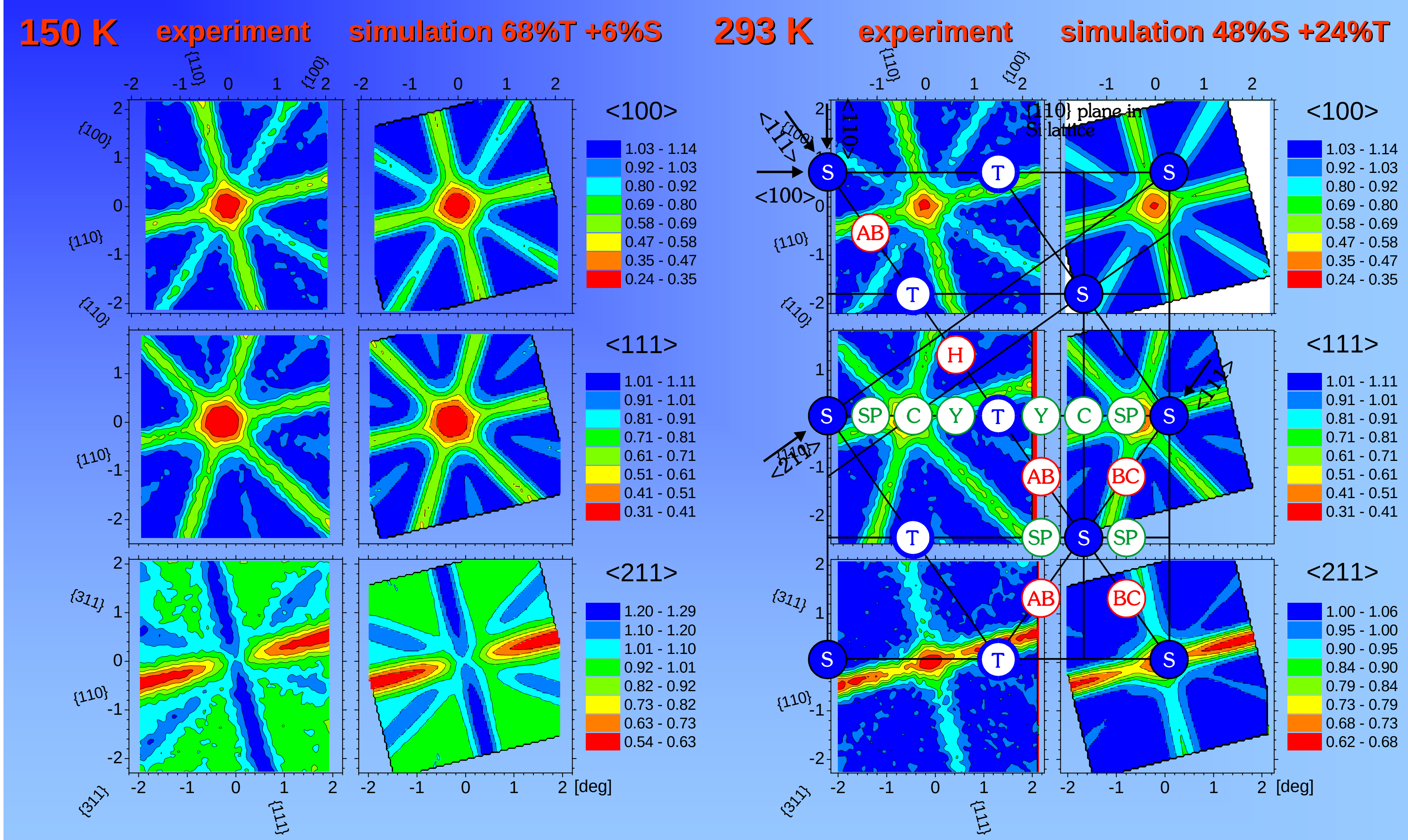
At much higher temperatures, Li^+ which was substitutional may leave this site again, leaving behind a III-vacancy

^{8}Li (838 ms)→β⁻2α decay for α emission channeling



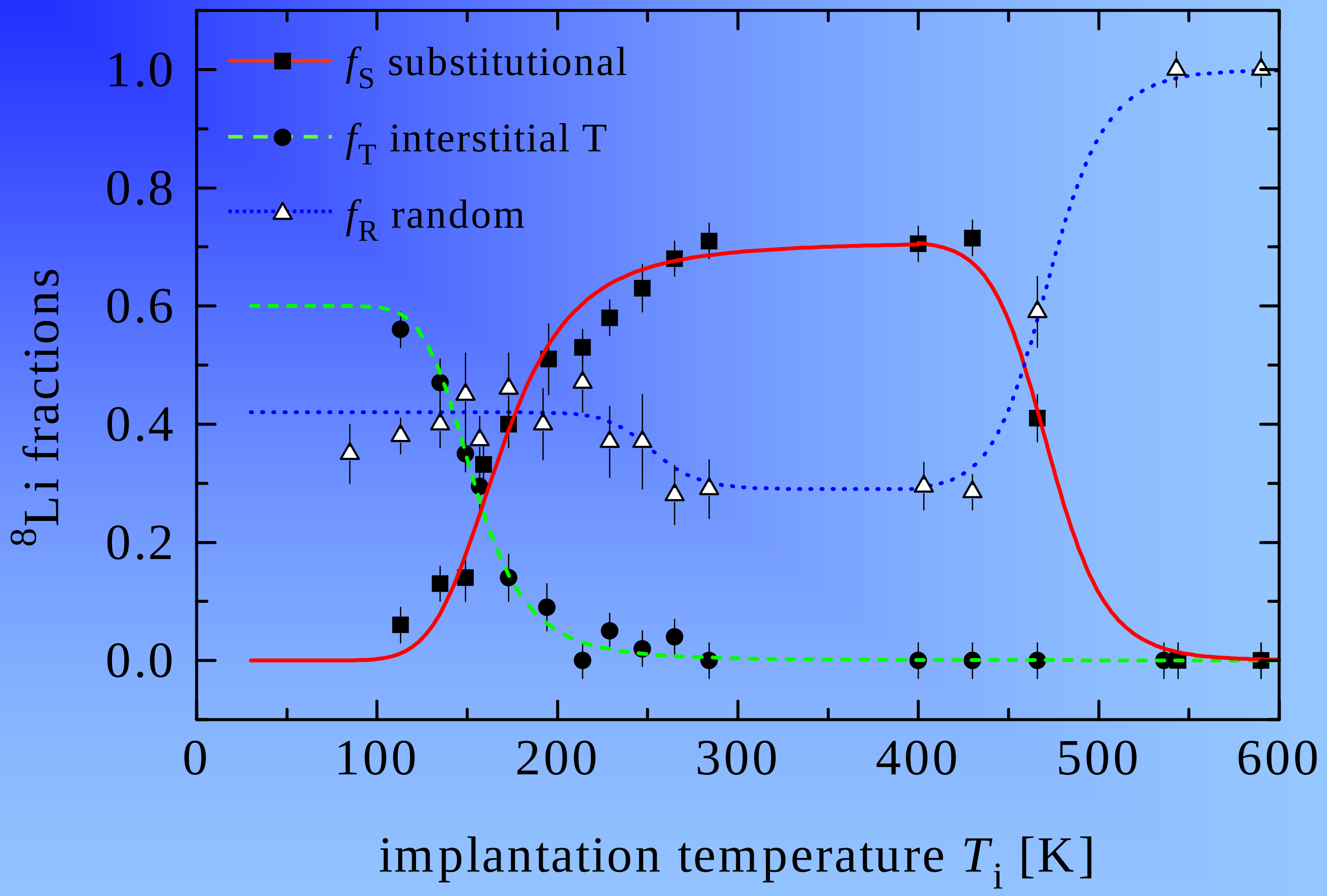
α particles from ^{8}Li β-delayed α decay make it a very efficient emission channeling probe nucleus

α emission channeling: lattice site changes of ^{8}Li (838 ms)→β⁻2α in GaAs



Site change of majority of Li from tetrahedral interstitial to substitutional Ga sites clearly visible from the <211> patterns

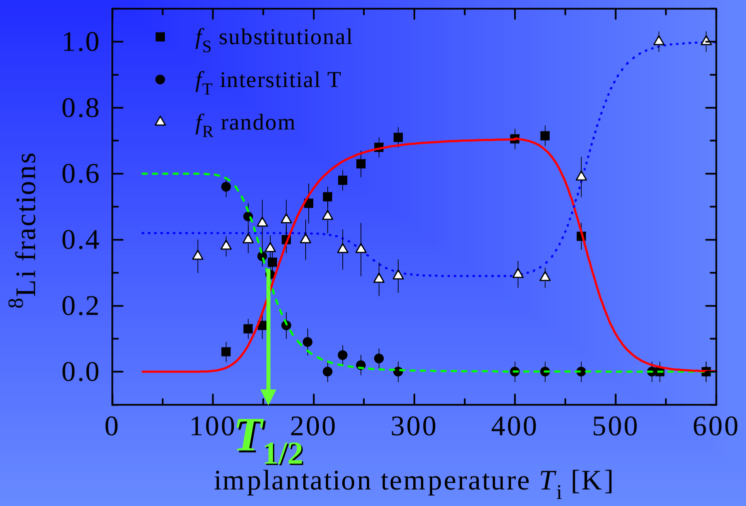
Emission channeling lattice site changes of ^{8}Li (838 ms) in InSb (1)



In InSb the site change of ^{8}Li from tetrahedral interstitial to substitutional In occurs between 100-200 K

U. Wahl, Physics Reports 280 (1997) 145

Emission channeling lattice site changes of ^{8}Li (838 ms) in InSb (2)



$Ze = -1, -2, -3$ charge of vacancy

R_C Coulomb capture radius $R_C(Z) = \frac{Ze^2}{\varepsilon k_B T}$ ≈ 48-174 Å

τ = 1208 ms radioactive lifetime of ^{8}Li

$\langle r_i^3 \rangle \approx 61$ Å mean cube distance of $^8Li^+$ and vacancy (estimated from MARLOWE or TRIM simulations)

D_0 entropy constant, estimated from $D_0 \approx \frac{l^2}{6} N_{NN} \nu_0$

Migration energy E_M of interstitial Li can be estimated by

$$E_M = kT_{1/2} \ln\left[\frac{3 \ln 2 R_C(Z) D_0 \tau}{\langle r_i^3 \rangle}\right] \approx 24 - 27\, kT_{1/2}$$

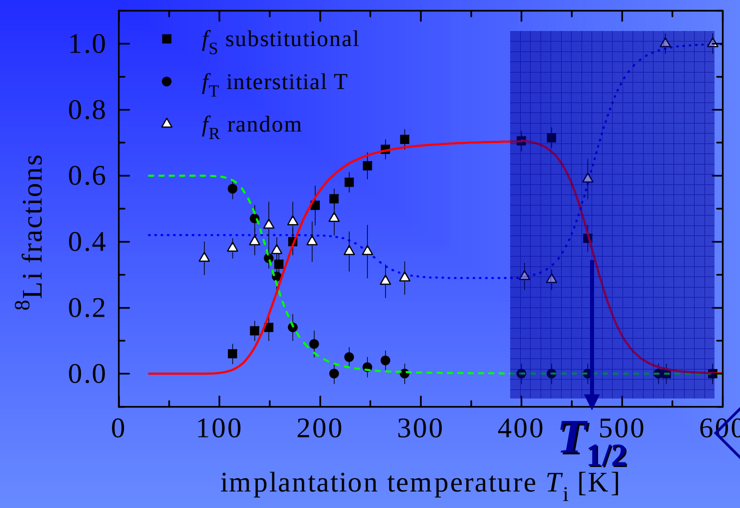
electric field drift model

For InSb: $T_{1/2} \approx 150$ K $\Rightarrow E_M \approx 0.31\text{-}0.35$ eV

In comparison: activation energy from macroscopic diffusion experiments $E_D = 0.28$ eV [Takabatake 1966]

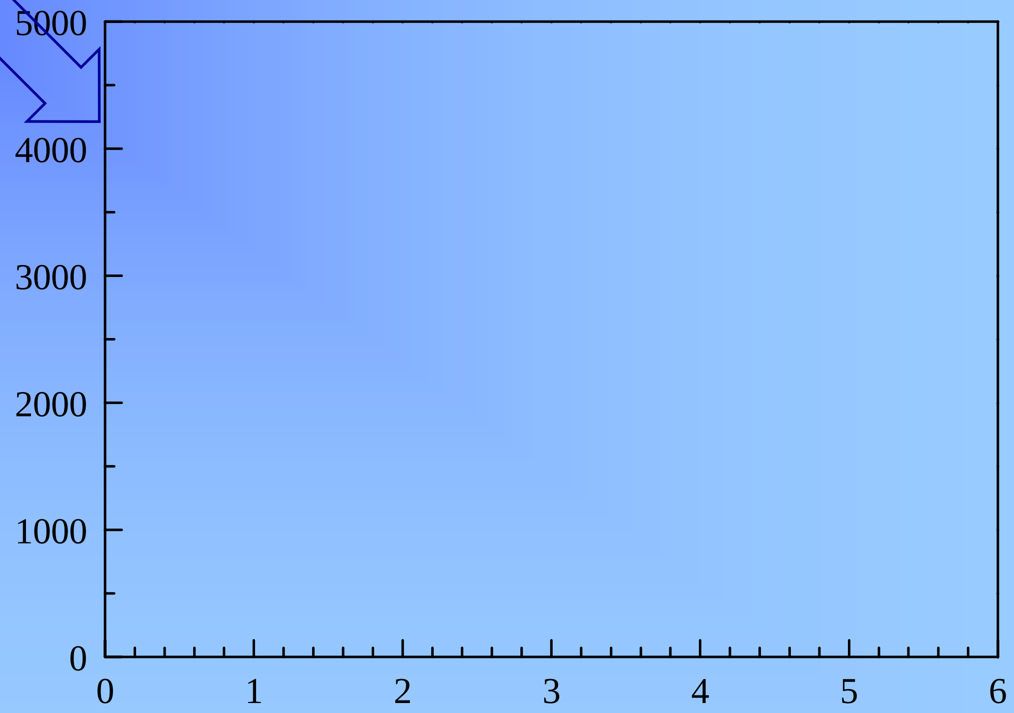
U. Wahl, Physics Reports 280 (1997) 145

Emission channeling lattice site changes of ^{8}Li (838 ms) in InSb (3)



Temperature regime where Li leaves substitutional In sites and long-range diffusion sets in

Shift of α energy spectrum shows Li diffuses to the surface of the sample



Activation energy E_A for dissociation of substitutional Li_{In} can be estimated from

$$E_A = kT_{1/2} \ln\left[\nu_0 \tau / \ln 2\right]$$

For $\nu_0 \approx 10^{13}$ s^{-1}

$\Rightarrow E_A \approx 1.1\text{-}1.2$ eV

U. Wahl, Physics Reports 280 (1997) 145

Li in semiconductors: diffusion data from emission channeling and other methods

Semi-conductor	T_{melt} [K]	T(T→S) [K]	E_M(Li) [eV]	E_D(Li) fast [eV]	T(S→R) [K]	E_A(S→R) [eV]	E_D(Li) slow [eV]
Si	1685	260-345	**0.56-0.65**	**0.655**	(>700)		
SiC	3100	(>850)	**(>1.8-2.0)**	**1.7-2.1**			
Ge	1210			**0.512**			
AlN	3100	650-750	**1.7**		(>800)		
GaN	2800	650-750	**1.7**	***1.4-1.55***	(>800)		
GaP	1730	495-530	**1.14-1.19**		(>700)		
GaAs	1513	240-270	**0.58-0.60**	**0.67**	(>300)		**1.2**
GaSb	985			**0.7**			**1.9**
InP	1335	360-405	**0.87-0.91**		(>600)		
InSb	800	155-180	**0.37-0.39**	**0.28**	450-500	**1.1-1.2**	
CdTe	1370	130-190	**0.33-0.37**		450-470	**1.2**	
ZnO	2250			***0.58-0.64***			
ZnSe	1790	200-230	**0.47-0.59**	**0.49**	500-550	**1.3-1.4**	
ZnTe	1510	150-180	**0.34-0.38**	**0.78**	450-470	**1.2**	**1.22**

Emission channeling data has helped in clarifying the diffusion mechanisms of Li.

U. Wahl, Physics Reports 280 (1997) 145

Examples for experimental results

- ❑ α emission channeling:
lattice location of ^{8}Li in semiconductors

(diffusion processes of Li in semiconductors, Li-defect interactions)

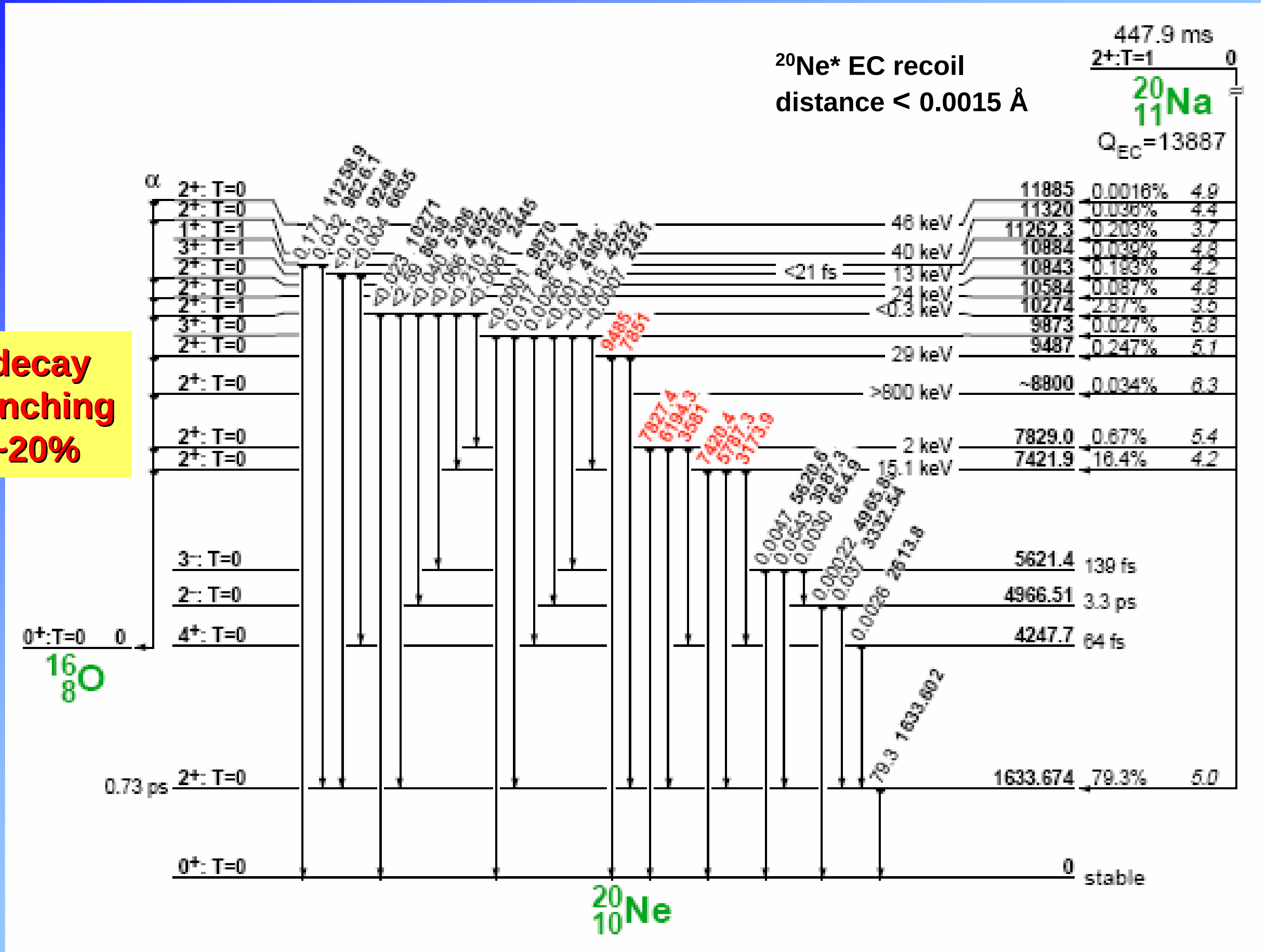
- ❑ β^{-} emission channeling:
lattice location of ^{24}Na in ZnO

(interstitial Na_i donors *vs* substitutional Na_{Zn} acceptors)

- ❑ β^{-} and CE emission channeling:
lattice location of rare earths (REs) in GaN

(REs as optical or magnetic dopants)

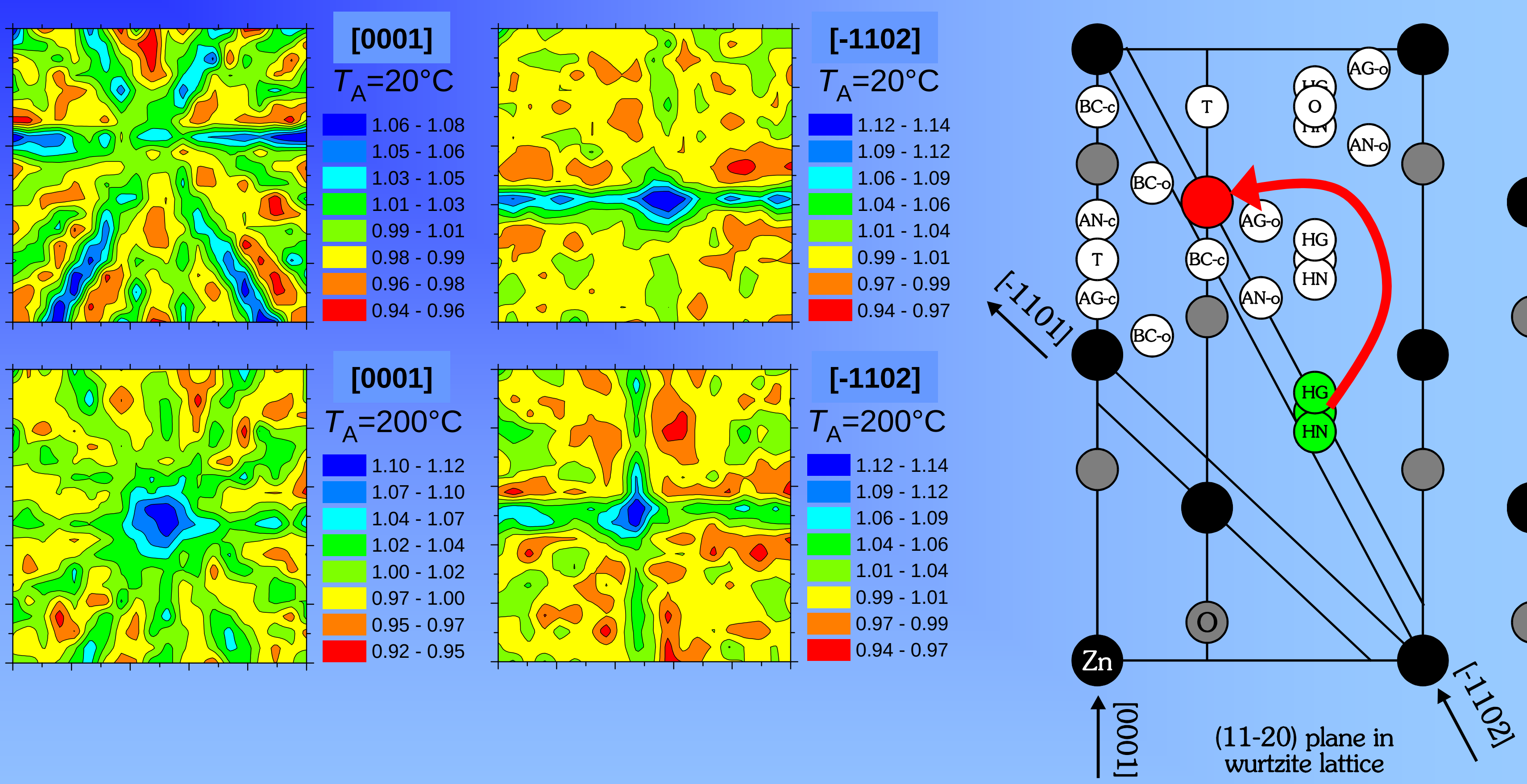
^{20}Na (448 ms)→EC⁻α decay for α emission channeling



α decay branching ~20%

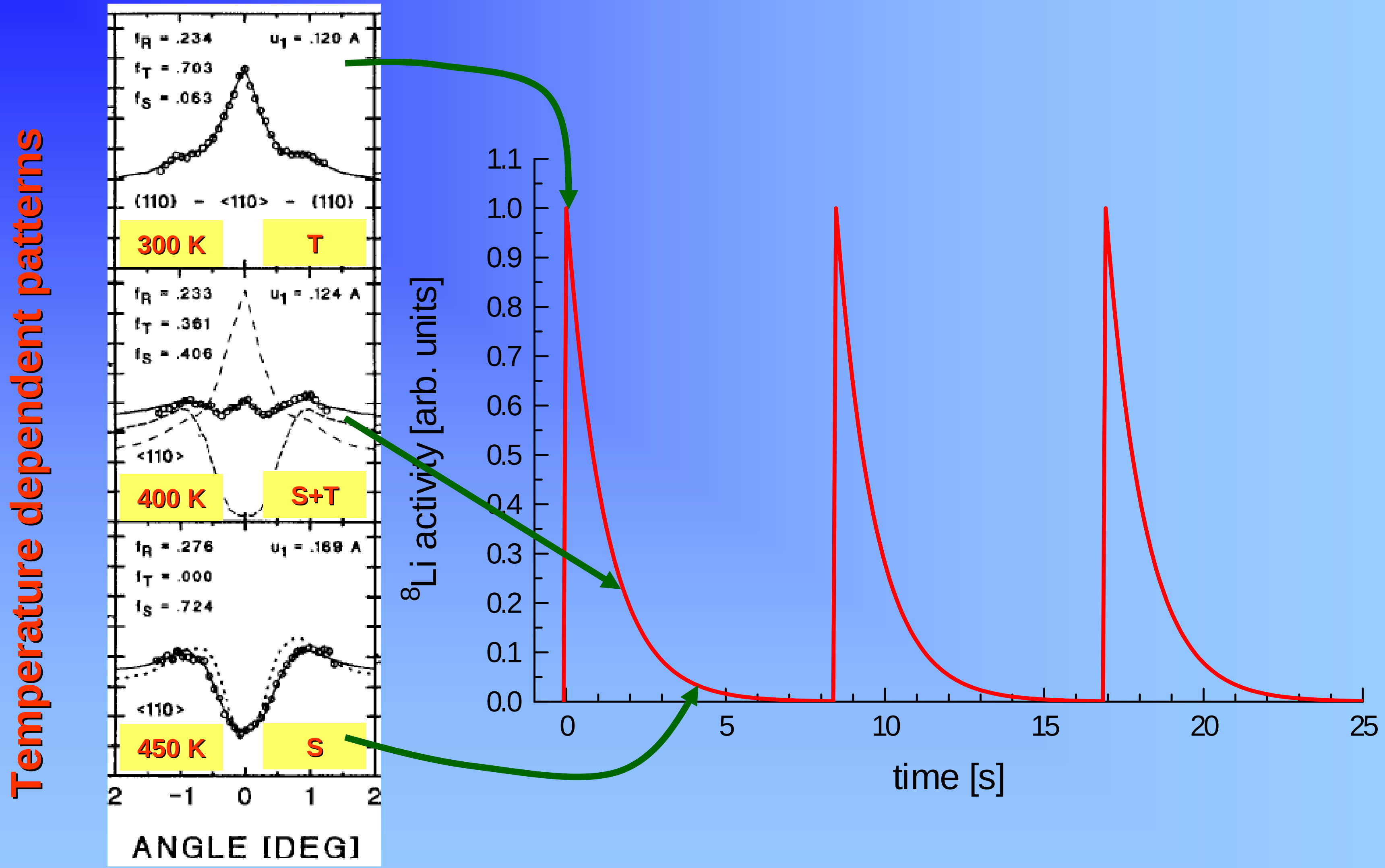
^{20}Na is a very interesting α emission channeling probe but experiments at ISOLDE were unsuccessful due to insufficient isotope yield ⇒ used ^{24}Na β⁻ instead

Emission channeling from ^{24}Na (14.9 h) β^- in ZnO



- as-implanted the majority of ^{24}Na sits on interstitial H or O_i sites in ZnO
- upon annealing at 200°C it changes to substitutional S_{Zn} sites

Concept for time-dependent emission channeling using pulsed beams



Measuring time dependent channeling patterns from ^{8}Li or ^{20}Na should allow to study in detail the kinetics of lattice site changes
(requires pulsed beams and good statistics in all time windows)

Examples for experimental results

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  lattice location of ^{8}Li in semiconductors

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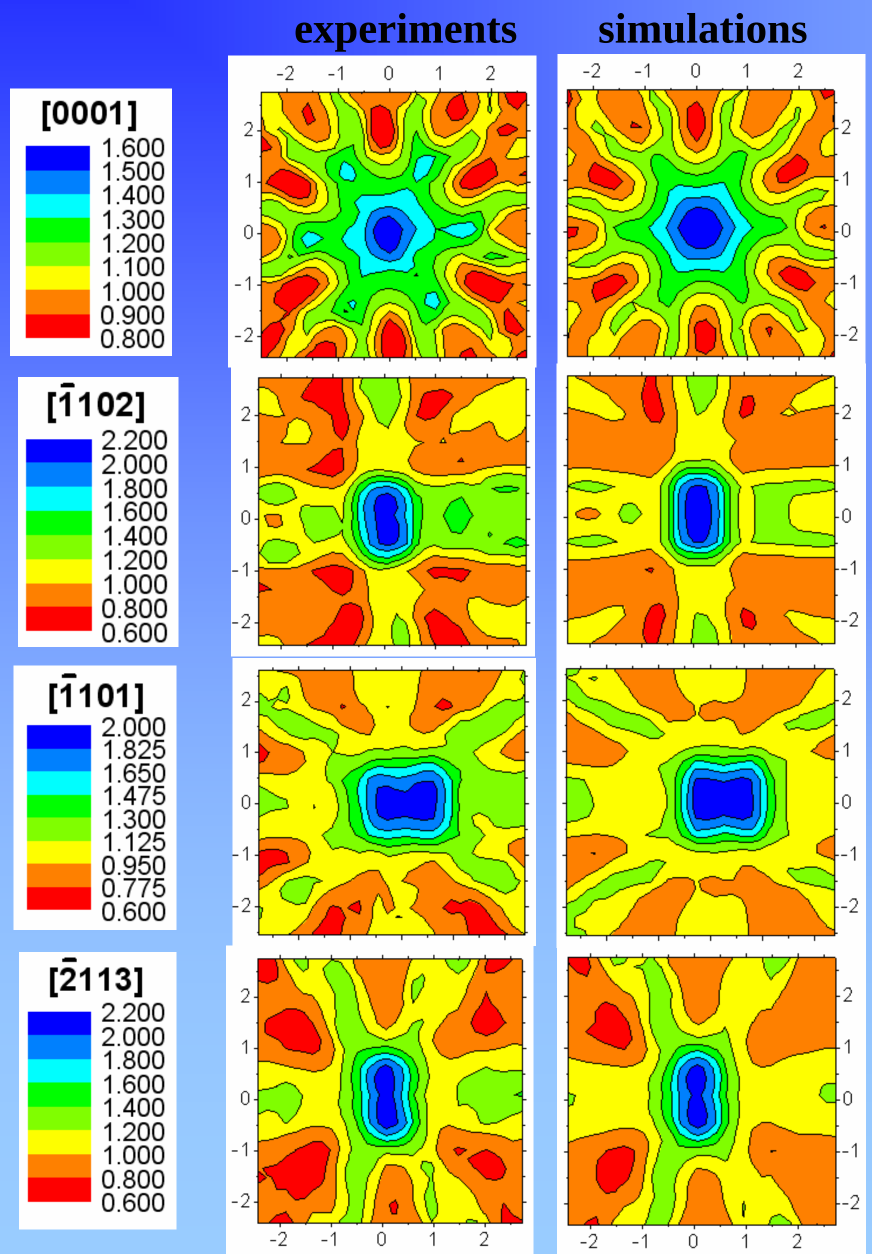
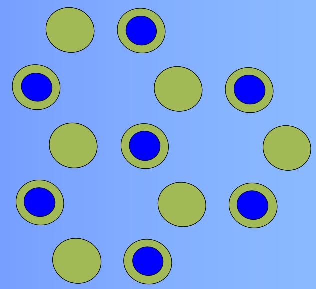
  lattice location of rare earths (REs) in GaN

  (REs as optical or magnetic dopants)

Conversion Electron (CE) emission channeling

$^{170}Lu \rightarrow {}^{170*}Yb$ in GaN T_A=600°C



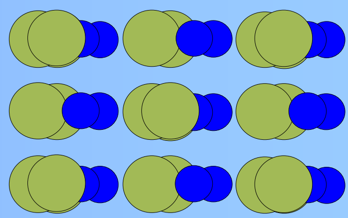


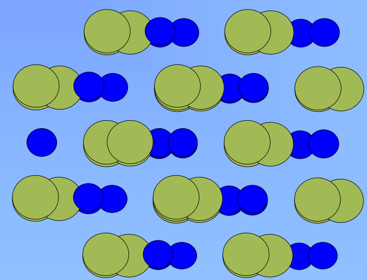
72% along *c*-axis atom rows
28% random

$u_{\perp[0001]}$ = 0.13 Å

81% along Ga atom rows

$u_{\perp[-1102]}$ = 0.16 Å



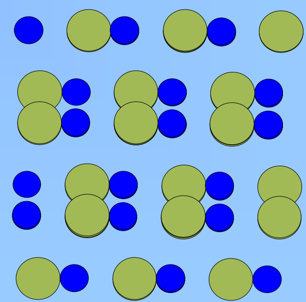


76% along Ga atom rows

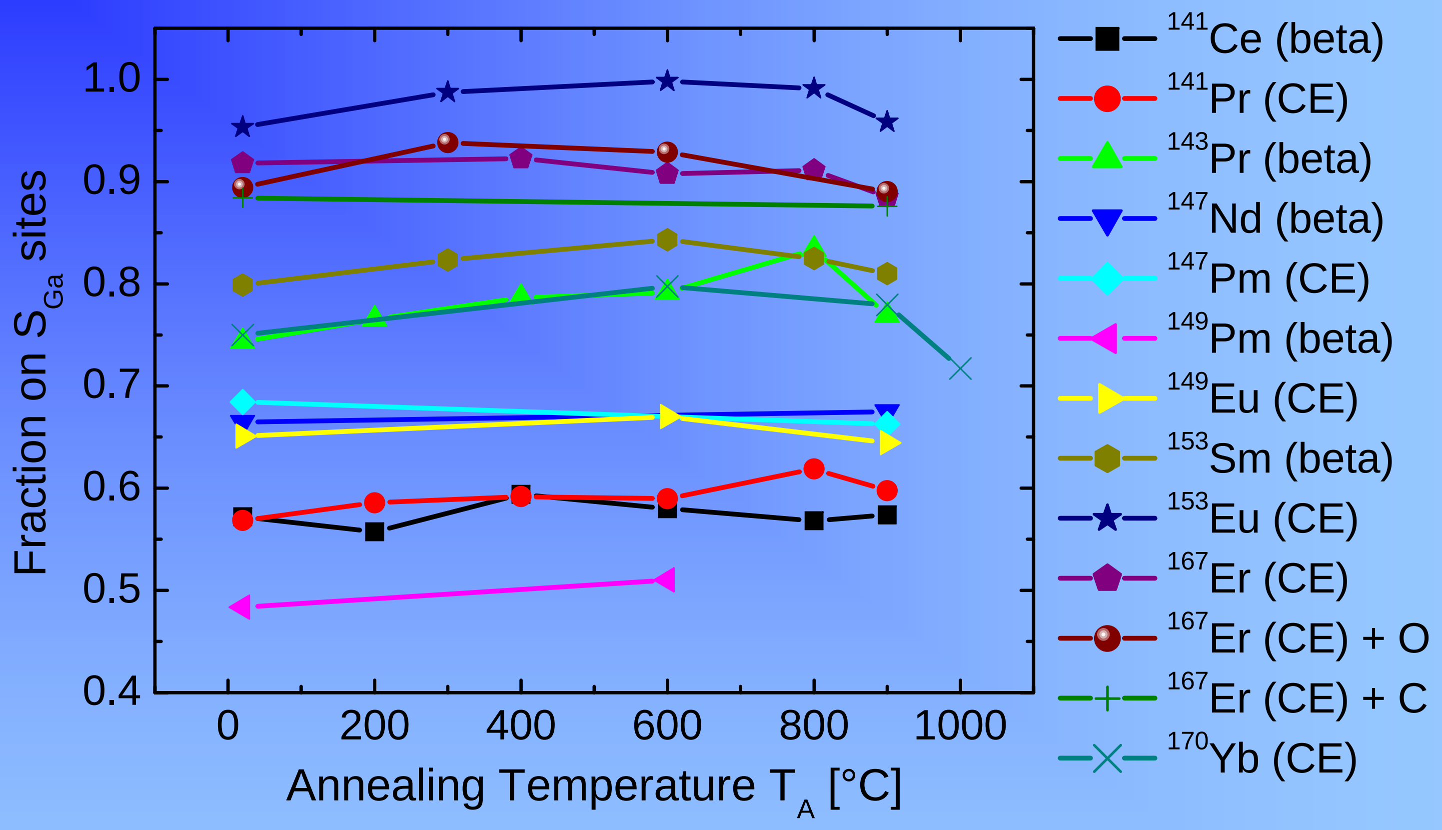
$u_{\perp[-1101]}$ = 0.14 Å

91% along Ga atom rows

$u_{\perp[-2113]}$ = 0.18 Å



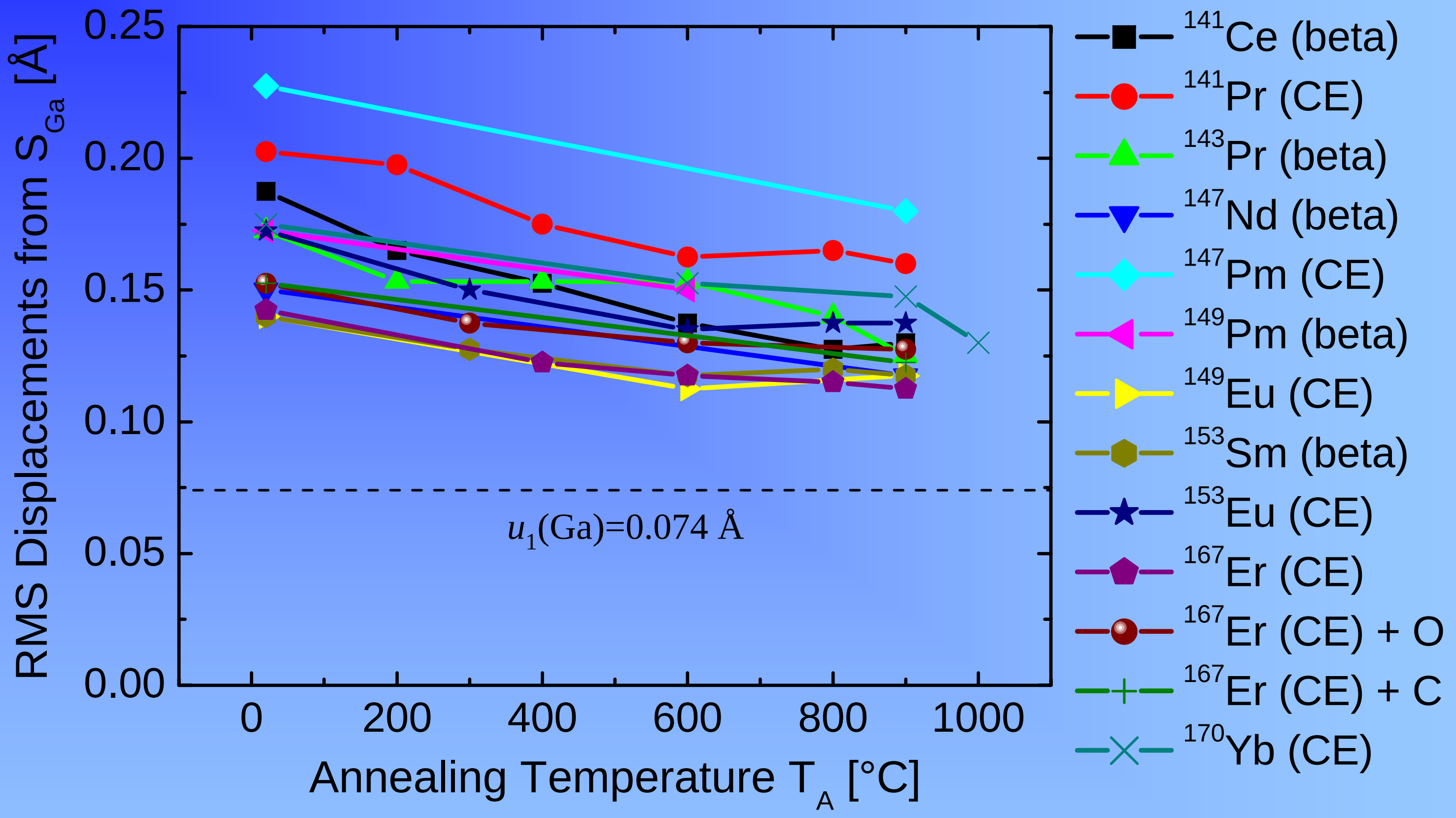
Rare earths in GaN: fractions on substitutional Ga sites



- large fractions of RE on S_{Ga} sites
- annealing hardly influences fractions
- actual differences between various REs possibly smaller (varying sample quality, **electron scattering background** etc.)

B. De Vries, PhD thesis KULeuven (2006)

Rare earths in GaN:
rms displacements from substitutional Ga sites



- displacements larger than thermal vibration
- decrease after annealing
- the only visible effect of annealing in GaN at these low doses

B. De Vries, PhD thesis KULeuven (2006)

Short-lived α emitters of interest for emission channeling

Some α-active isotopes which are of interest for future emission channeling studies in semiconductor physics

Isotope	$t_{1/2}$	α yield per decay	E_α (MeV)	ISOLDE 2 yield (ions s^{-1} μA^{-1})	PSB ISOLDE yield (ions s^{-1} μA^{-1})	Target material
^{8}Li	0.838 s	2.0	ca. 1–5.5	3.9×10^{8}	1.1×10^{9}	Ta
^{8}B	0.769 s	2.0	ca. 1–5.5	Not available	Not available	
^{20}Na	0.446 s	0.21	2.148, 4.438	Not available	~~9.8×10^{8}~~	Al
^{150}Dy	7.17 min	0.31	4.23	2.4×10^{9}		Ta
^{151}Dy	17 min	0.06	4.07	7.3×10^{9}		Ta
^{151}Ho	35.6 s	0.10	4.52	1.0×10^{8}		Ta
^{152}Er	9.8 s	0.10	4.80	7.0×10^{7}		Ta
^{153}Er	36 s	0.38	4.67	7.1×10^{8}		Ta
^{153}Tm	1.58s	1.0	5.10	1.0×10^{7}		Ta
^{154}Tm	3.0s	1.0	5.03	1.1×10^{8}		Ta
^{155}Tm	39s	1.0	4.45	4.0×10^{8}		Ta

- of those only ^{8}Li was used at ISOLDE
- on-line emission channeling experiments require long beam times, well-focused beams, good yields and fast detection systems

U. Wahl, Physics Reports 280 (1997) 145

Conclusions

- Emission channeling is a sensitive technique that can give direct information on the lattice sites of impurities in single crystals with ~0.1 Å precision (in favourable cases ~0.01 Å).
- Emission channeling experiments significantly contributed to our knowledge on the lattice sites of impurities in semiconductors, metals and oxides, in many cases yielding results that could not have been obtained by other methods.
- A MYRRHA-based ISOL facility offers opportunities for emission channeling experiments, particularly for ^{8}Li, ^{20}Na, and rare earth α emitters.

Thanks to...

Emission Channeling Lattice Location Experiments with Short-Lived Isotopes

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